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Seattle Distribution System Corrosion Control Study. Volume III: Potential for Drinking Water Contamination from Tin/Antimony Solder

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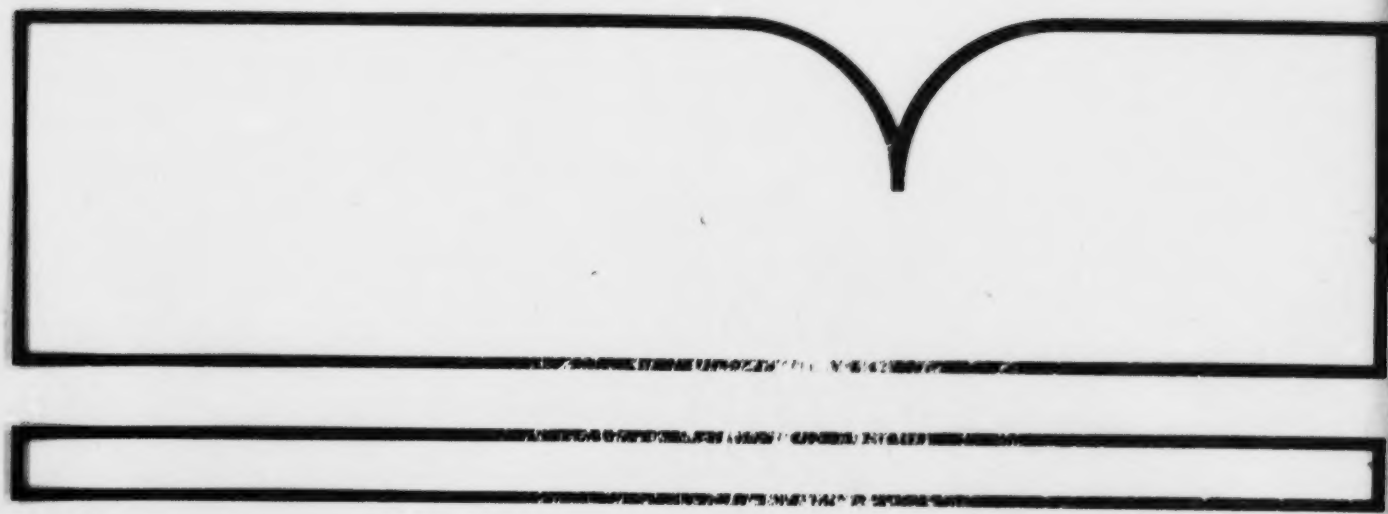

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SEATTLE DISTRIBUTION SYSTEM CORROSION CONTROL STUDY

Volume III. Potential for Drinking Water Contamination from Tin/Antimony Solder

by

Carlos E. Herrera
Gregory J. Kirmeyer
Brian P. Hoyt
Seattle Water Department
Seattle, Washington 98144

Contract No. R 806686 010

Project Officer

Marvin C. Gardels
Drinking Water Research Division
Municipal Environmental Research Laboratory
Cincinnati, Ohio 45268

MUNICIPAL ENVIRONMENTAL RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
CINCINNATI, OHIO 45268

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16. ABSTRACT

This study was conducted to evaluate the potential for leaching of metals into drinking water from tin/antimony solder. The study consisted of five research phases: (1) A theoretical investigation of the corrosion products formed from the solder was conducted using pe-pH diagrams and galvanic coupling relationships; (2) static laboratory metal leaching tests were performed to determine the extent of antimony leaching from the solder; (3) field water samples were taken from buildings which contained tin/antimony from tin/antimony soldered plumbing; (4) continuous flow metal leaching tests compared metal leaching from tin/antimony solder with lead/tin solder under three water treatments - lime plus sodium carbonate, lime plus sodium bicarbonate, and lime plus bicarbonate plus silicate; (5) corrosion films that formed on both tin/antimony and lead/tin solder were analyzed by Scanning Electron Microscopy and X-ray diffraction.

The results indicate that increases in metals concentrations resulting from the corrosion of tin/antimony solder are minimal. Metal leaching from the solder is inhibited by the sacrificial corrosion of tin and the formation of a stable double layer tin passivation film (SnO and SnO2). Side-by-side tests comparing metal leaching from tin/antimony and lead/tin solder show substantial reductions in standing water lead concentrations when tin/antimony solder is used.

The use of tin/antimony solder in place of lead-based solders should result in reduced human exposure to lead from drinking water with only minimal increases in antimony exposure.

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FOREWORD

The U.S. Environmental Protection Agency was created because of increasing public and government concern about the dangers of pollution to the health and welfare of the American people. Noxious air, foul water, and spoiled land are tragic testimonies to the deterioration of our natural environment. The complexity of that environment and the interplay of its components require a concentrated and integrated attack on the problem.

Research and development is that necessary first step in problem solution; it involves defining the problem, measuring its impact, and searching for solutions. The Municipal Environmental Research Laboratory develops new and improved technology and systems to prevent, treat, and manage wastewater and solid and hazardous waste pollutant discharges from municipal and community sources, to preserve and treat public drinking water supplies, and to minimize the adverse economic, social, health, and aesthetic effects of pollution. This publication is one of the products of that research and provides a most vital communications link between the researcher and the user community.

This report presents the results and conclusions from metal leaching research on the corrosion of solders in an aggressive water supply. The data support the conclusion that reductions in drinking water metals levels can be achieved by proper material selection.

Francis T. Mayo, Director
Municipal Environmental Research
Laboratory

ABSTRACT

A study was conducted in Seattle, Washington, to evaluate the potential for leaching of metals into drinking water from tin/antimony solder. The study consisted of five research phases: (1) A theoretical investigation of the corrosion products formed from the solder was conducted using pe-pH diagrams and galvanic coupling relationships. (2) Static laboratory metal leaching tests were performed to determine the extent of antimony leaching from the solder. (3) Field water samples were taken from buildings which contained tin/antimony soldered plumbing. (4) Continuous flow metal leaching tests compared metal leaching from tin/antimony solder with lead/tin solder under three water treatments - lime plus sodium carbonate, lime plus sodium bicarbonate, and lime plus sodium bicarbonate plus silicate. This evaluation method was developed by the Seattle Water Department for this study. (5) Corrosion films that formed on both tin/antimony and lead/tin solder were analyzed by Scanning Electron Microscopy and X-ray diffraction.

The results indicate that increases in metals concentrations resulting from the corrosion of tin/antimony solder are minimal. Metal leaching from the solder is inhibited by the sacrificial corrosion of tin and the formation of a stable double layer tin passivation film (SnO and SnO_2). Side-by-side tests comparing metal leaching from tin/antimony and lead/tin solder show substantial reductions in standing water lead concentrations when tin/antimony solder is used.

These tests affirm an earlier Seattle Water Department recommendation to eliminate the use of lead-based solders in Seattle's domestic water plumbing. The use of tin/antimony solder in place of lead-based solders should result in reduced human exposure to lead from drinking water with only minimal increases in antimony exposure.

This report was submitted in fulfillment of Grant No. R 806686 010 by the Seattle Water Department under the sponsorship of the U.S. Environmental Protection Agency. This report covers the period from June 1979 to February 1981, and work was completed as of February 1981.

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ABBREVIATIONS AND SYMBOLS

ABBREVIATIONS

A.A.S.	-- atomic absorption spectrophotometry
A/C	-- asbestos cement
aq	-- aqueous species
C	-- solid species
^{o}C	-- degrees Celsius
cm	-- centimeter
cm^2	-- square centimeters
CPVC	-- chlorinated polyvinyl chloride
EDAX	-- energy dispersive x-ray detector
E_H	-- standard equilibrium potential
EPA	-- U.S. Environmental Protection Agency
F	-- Faraday constant
g	-- gaseous species
ΔG^{o}	-- standard free energy of formation
(Halogen + $SO_4^{=}$)/Alk	-- (halogen + sulfate)/ alkalinity ratio
K	-- equilibrium constant
kg	-- kilogram
L	-- liter
L/s	-- liter per second
m	-- meter
m^3	-- cubic meter
m^3/d	-- cubic meter per day
mg	-- milligram
mg/L	-- milligram per liter
m/s	-- meter per second
MCL	-- maximum contaminant level
NA	-- not analyzed
OX	-- oxidized species
PB	-- polybutylene
ppm	-- parts per million
R	-- gas constant
RED	-- reduced species
SEM	-- scanning electron microscopy
SWD	-- Seattle Water Department
T	-- temperature
50/50 Pb/Sn	-- 50% lead, 50% tin solder
95/5 Sn/Sb	-- 95% tin, 5% antimony solder
µg/L	-- micrograms per liter
µm	-- micrometers
µmhos	-- micromhos

SYMBOLS

$CaCO_3$	-- calcium carbonate
$Ca(OH)_2$	-- calcium hydroxide (lime)
Cd	-- cadmium
CO_2	-- carbon dioxide
Cu	-- copper
e^-	-- electron
Fe	-- iron
H^+	-- hydrogen ion
H_2O	-- water
HNO_3^-	-- nitric acid
$HSbO_2$	-- antimonious acid
Na_2CO_3	-- sodium carbonate (soda ash)
$NaHCO_3$	-- sodium bicarbonate
$NaO-SiO_2$	-- sodium silicate
Pb	-- lead
Pd	-- palladium
pe	-- -log $\{e^-\}$
pH	-- -log $\{H^+\}$
pH_2	-- partial pressure of hydrogen
pO_2	-- partial pressure of oxygen
Sb	-- antimony
Sb_2O_3	-- antimony trioxide
Sb_2O_4	-- antimony tetroxide
Sb_2O_5	-- antimony pentoxide
SbO^+	-- antymonyl ion
SbO_2^-	-- antimonite ion
$SO_4^{=}$	-- sulfate

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SECTION 1

INTRODUCTION

SCOPE

This report is Volume III of a seven-volume series presenting results of research conducted under the Seattle Distribution System Corrosion Control Study. This volume evaluates the potential of drinking water contamination from the corrosion of tin/antimony solder. Volume I presents pilot test results and recommends specific chemical treatment for internal corrosion control in the Cedar water distribution system. Volume II presents the results of a pilot plant study for internal corrosion control in the Tolt water distribution system. Volume IV evaluates the effectiveness of point-of-use corrosion inhibitor systems. Volume V presents the results of a pilot plant study conducted to determine the effects of chemical treatment on old galvanized pipe. Volume VI presents a laboratory electrochemical technique for corrosion rate determinations in drinking water. And Volume VII addresses the counteractive effects of disinfection (chlorination) and corrosion control (pH/alkalinity adjustment).

BACKGROUND INFORMATION

Seattle Service Area

The Seattle Water Department (SWD) service area includes Seattle proper and also most of the metropolitan area. It extends from Edmonds on the north to just south of Des Moines, and easterly from Puget Sound to Lake Sammamish at the foot of the Cascade range. Over 1 million persons are served directly or indirectly by the SWD, which represents about 83% of the population of King County. Average total water consumption is 609,000 cubic meters per day (m^3/d), and summer peak day use has reached 1.3 million m^3/d. Approximately two-thirds of this water is sold directly to the SWD retail customers, and the remaining portion is delivered at wholesale rates to suburban water districts and municipalities, who in turn sell it to their direct service customers.

Water Sources

The primary sources of Seattle water are the Cedar and Tolt rivers on the western slope of the Cascade Mountains in eastern King County. These two rivers furnish high quality mountain water of unusual clarity. The Cedar River water supply system, which has been in operation since 1901, serves the southern region of Seattle and parts of South King County. The

Tolt, which was added to the City system in 1964, generally serves the area north of the Lake Washington Ship Canal and the water purveyors on the east side of Lake Washington. These two sources feed the distribution system from opposite ends, a feature that affords excellent hydraulic characteristics. Figure 1 illustrates the major facilities of the system.

Water Treatment

Currently, the treatment of both water supplies consists of intake screening, chlorination and fluoridation. Prior to distribution, the water is rechlorinated as it leaves the open in-town reservoirs to assure the free chlorine residual necessary to maintain adequate levels of disinfection. Compared to most other major water utilities in this country, the present treatment is minimal. Many utilities are required to extensively treat their water with coagulation and filtration techniques to render it safe to drink. The SWD's emphasis on watershed protection, together with the large storage reservoir system to meet demands during periods of high turbidity, has enabled Seattle to meet National Interim Primary Drinking Water Regulations[1] without major modifications to the existing facilities.

Existing Facilities on the Cedar and Tolt River Systems

Cedar --

The Cedar River system originates in a 370-square-kilometer (km^2) watershed located in eastern King County. Eighty percent of the ownership of the watershed is held by the City of Seattle. The remaining 20% is divided between the United States Forest Service and the Weyerhaeuser Company. From the watershed, the river flows into Morse Lake, which is located on the site of the naturally occurring inundated Cedar Lake. Water is normally released from Morse Lake back into the natural river channel through a power generating station located at Cedar Falls. Water flows approximately 18 kilometers (km) before being diverted at the Landsburg intake into a 2.44-meter (m) diameter tunnel for transmission to the Seattle area. At Landsburg, the water is treated by intake screening, chlorination, and fluoridation. After treatment, the water can be directed into the distribution system or into Lake Youngs, a large storage reservoir located just southeast of Seattle. Water is again rechlorinated at the outlet from Lake Youngs.

Tolt --

The Tolt River system originates in a 54-km^2 area watershed, which is narrow and steep and has been prone to slide activity. Ownership in the watershed is about equally divided between the Weyerhaeuser Company, the U.S. Forest Service, and the SWD. Ten major streams feed the South Fork Tolt Reservoir, a 69-million-cubic-meter (m^3) manmade storage reservoir with a maximum pool elevation of 536 m. The water enters a 84-centimeter (cm) diameter, 7.7-km pipeline through one of three intakes located at different depths in the reservoir. Upon discharge from the pipeline, water enters the pressure regulating basin, a 108,000-m^3 reservoir at an elevation of 231 m. The purpose of the pipeline and the regulating basin is to dissipate 305 m of head. Water then enters a 168-cm-diameter transmission line, where it is chlorinated, fluoridated, and subsequently discharged into the Seattle distribution system.

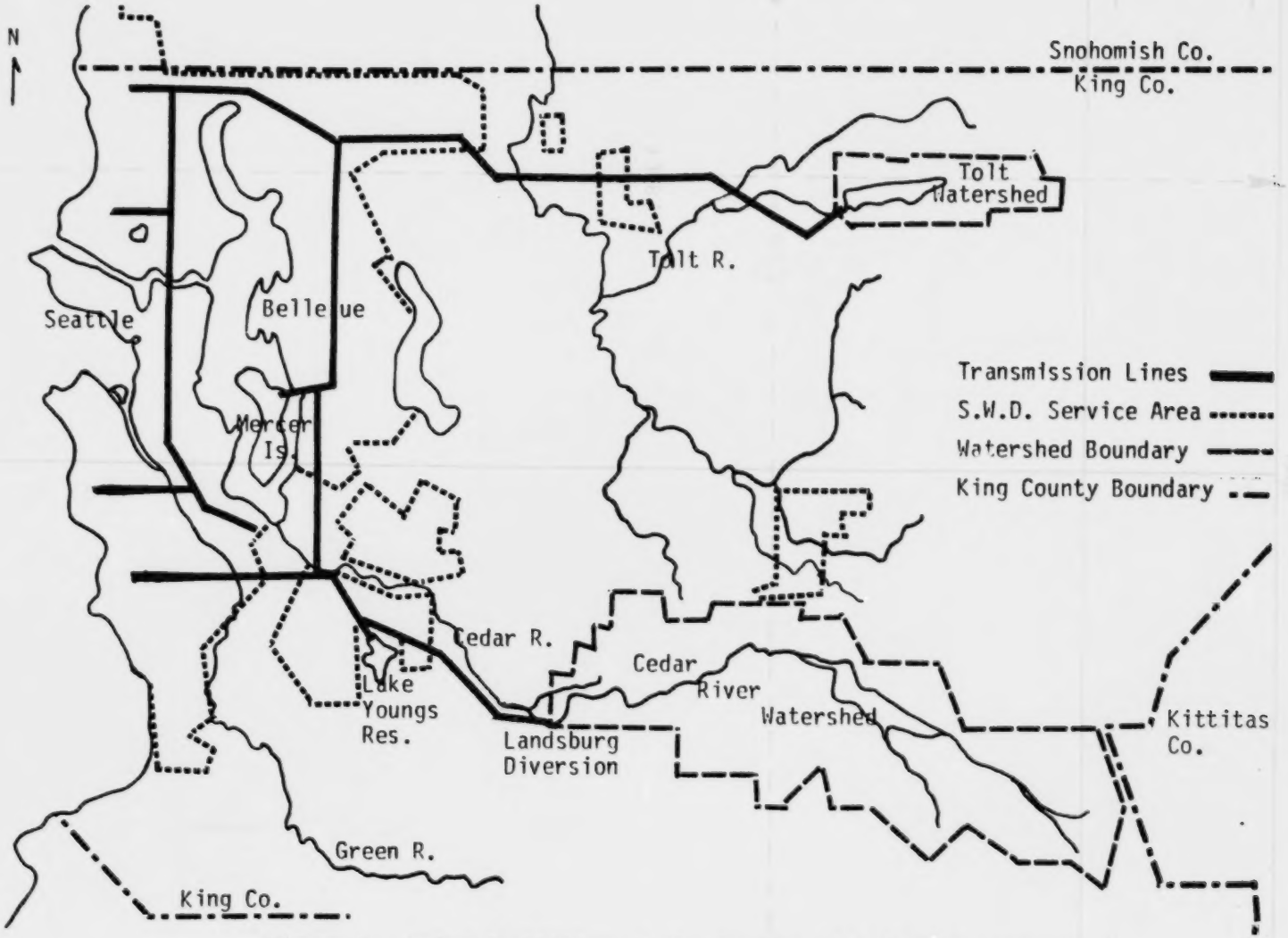

Figure 1. Major facilities of the Seattle distribution system.

Causes of Corrosion

Although the quality of both the Cedar and Tolt water supplies is quite high, both are corrosive to premise plumbing systems. The corrosiveness of the water results from several related factors, which include:

Acidity as indicated by low pH; after chlorination and fluoridation, pH is 6.8 to 7.2 for the Cedar supply and 5.8 to 6.2 for the Tolt supply;

A dissolved oxygen concentration that is at saturated conditions;

Insufficient calcium and bicarbonate alkalinity in the water to form protective calcium carbonate films on pipe surfaces;

A relatively high [halogen + sulfate] / alkalinity ratio [(Halogen + $SO_4^=$) / Alk] of 0.5 to 0.8 for the Cedar and 2.5 for the Tolt, which is an indicator of conditions favorable to pitting corrosion.

In 1970, three factors combined to intensify the corrosiveness of both water supplies: (1) To decrease the occurrence of positive bacteriological samples within the distribution system, the chlorine dosage at the open reservoir outlets was increased. (2) At the request of the U.S. Public Health Service, ammoniation of the water supply was stopped to enable a free chlorine residual to be maintained throughout the distribution system. This change from combined chlorination to free chlorination was implemented to provide quicker, more effective disinfection of the unfiltered water supply. (3) Based on a vote of the Seattle citizens in 1968, fluoridation with hydrofluosilicic acid began in 1970.

Internal Corrosion Problem

Water quality deterioration in home and building plumbing systems is a major concern of the SWD. Through several projects that have examined the corrosivity of its water supplies, it is now well documented that corrosion, although not a major concern in the distribution system, causes several problems in premise plumbing.[2] In addition to causing the obvious aesthetic problems of rusty water, red and blue staining and metallic tastes; plumbing corrosion is responsible for a sizable economic burden of Seattle homeowners and possibly some long-term health impacts to those with high environmental exposure to lead.

Aesthetics --

Customer complaints of rusty water, stained fixtures and metallic tastes are frequently received from within the water distribution system. This problem has been well documented by accurate complaint records and by a questionnaire survey conducted by the SWD in 1973. The survey, which was distributed to 15,000 customers (10% of all services within the direct service area), showed that 16.7% of customers in the Cedar water distribution system and 28.3% of customers in the Tolt water distribution system experienced corrosion-related problems.[3]

Economics --

Significant corrosion of piping occurs in the premise plumbing systems served by Cedar and Tolt water. The average estimated life span of hot water galvanized and copper pipes is approximately 35 years in the Cedar system and 25 years in the Tolt system. The forecasted 1978 average annual cost for maintaining serviceability in these pipes was estimated to be approximately $7.8 million.[4]

Health Impact --

Evidence gathered after 1972 demonstrates that the levels of lead (Pb), copper (Cu), and iron (Fe) often exceed Maximum Contaminant Levels (MCL's) in overnight standing Cedar tap water (Table 1).

Cadmium (Cd) and zinc (Zn) were also found to increase after overnight standing in home plumbing; however, they rarely exceeded their MCL's.

Although the health impact of overnight standing water metal levels is not an acute problem, it would be desirable to reduce exposure to metals, especially lead and cadmium.

Sources of Lead in Seattle Drinking Water

The sources of lead in Seattle drinking water include the lead solder which is used to join copper pipes and lead impurities in galvanized piping.[6] Fortunately, little, if any, lead water pipe has been used in the Seattle area.

The commonly used solder is 50% lead and 50% tin by weight (50:50). It has been used extensively throughout the service area and its use has increased rapidly in the past few years. Copper pipe with 50/50 lead/tin solder now accounts for about 95% of all new piping systems. The amount of solder exposed to the water is normally only a small amount but this depends on the care taken by the plumber who joins the pipe. Because of the high amount of lead in the 50/50 solder, pilot and field tests have documented increased lead concentrations in water after standing overnight in copper pipe systems joined with lead solder.[4, 5, 6]

Lead is also present in galvanized pipe, apparently imparted as an impurity during the galvanizing process. Most of the older plumbing pipes in the Seattle service area are galvanized steel. Several metal leaching surveys have documented increased lead concentrations in the water after standing overnight in galvanized plumbing systems.[4, 5, 6]

It should be emphasized that the samples mentioned above were taken from water that had been standing for an extended period of time in a home plumbing system. After an initial flushing of the system, little or no lead pick-up was observed. An analysis of results of several metal leaching surveys indicates that 13% of Tolt and 14% of the Cedar "standing" water samples exceeded the MCL of 0.05 milligrams per liter (mg/L) for lead. In a recent study, the National Academy of Sciences concluded that the present 0.05 mg/L limit may not provide an adequate margin of safety and recommended that EPA reduce the standard to one-half of its present level. The health

TABLE 1. PERCENTAGE OF SAMPLES EXCEEDING EPA MAXIMUM CONTAMINANT LEVELS

Survey	Water Supply	Primary Contaminants: Lead 0.05 mg/L R	Lead 0.05 mg/L S	Cadmium 0.01 mg/L R	Cadmium 0.01 mg/L S	Secondary Contaminants: Copper 1 mg/L R	Copper 1 mg/L S	Iron 0.3 mg/L R	Iron 0.3 mg/L S	Zinc 5 mg/L R	Zinc 5 mg/L S	Color 15 units R	Color 15 units S	Manganese 0.05 mg/L R	Manganese 0.05 mg/L S
SWMS* (1974-75)	Tolt	0	10	0	0	1	9	19	76	0	1	24	87	0	10
	Cedar	0	16	0	2	0	10	8	60	0	3	0	56	0	0
EPA† (1972)	Tolt	0	23	0	9	3	23	15	73	0	9	-	-	0	5
Kennedy‡ Engr/ SWD (1976)	Tolt	0	0	0	0	0	0	0	100	0	0	-	-	0	0
	Cedar	0	0	0	0	0	0	0	40	0	9	-	-	0	0
TOTAL	Tolt	0	13	0	2	2	11	17	76	0	3	24	87	0	7
	Cedar	0	14	0	2	0	10	11	59	0	3	9	56	0	0
	Tolt & Cedar	0	13	0	2	1	11	14	69	0	3	12	73	0	5

R = Running cold water samples.
S = Overnight standing cold water samples.
*Seattle Water Metals Survey.[5]
†Study of Corrosion Products in the Seattle Water Department Tolt Distribution System.[6]
‡Internal Corrosion Study.[4]

authorities have determined that the problem in Seattle is not an acute problem; however, they indicate that reduction of lead in "standing" water is a worthy goal and that it would have a positive impact on public health.

CORROSION CONTROL PLAN

Early Studies Alluding to Corrosion Problem

Studies conducted by the U.S. Environmental Protection Agency (EPA)[6] and a joint commission of health, regulatory and utility officials[5] documented significant water quality changes between the water sources and the customer's tap. These studies indicated that most of the changes were occurring between the meter and the customer's faucet and not in the utility distribution system. Early attempts by the SWD to reduce the corrosion problem through chemical additions to the more corrosive Tolt water were thwarted by actions of environmental groups. After those attempts, the Seattle City Council directed the SWD to retain a consulting engineering firm and to develop a comprehensive corrosion control plan for the entire water supply system. A study team consisting of an engineering consultant, a citizen advisory committee and the SWD was brought together and they began a three-year effort to develop a comprehensive corrosion control strategy.

From the corrosion studies,[4] it became evident that the serious corrosion problem had to be approached from two sides. The water chemistry had to be altered but in addition, the aspect of selection of corrosion resistant materials also had to be addressed. The SWD transmission and water distribution system was not suffering seriously from the effects of the corrosive water condition; this was primarily because of the selection of proper materials. The same approach also needed to be implemented for home and building plumbing systems. Thus, a two-phased approach was developed: (1) A water treatment program, and (2) an aggressive program relating to selection of corrosion resistant materials in home and building plumbing systems.

Water Treatment Program

The water treatment program that has been selected will increase the pH and alkalinity of the waters and will decrease the (Halogen + $SO_4^=$)/Alk ratio. The present and proposed water quality characteristics are presented in Table 2.

TABLE 2. PRESENT AND PROPOSED WATER QUALITY CHARACTERISTICS

	Present Quality of Chlorinated and Fluoridated Water		Proposed Water Quality After Corrosion Treatment	
Parameter	Cedar	Tolt	Cedar	Tolt
pH (pH units)	6.8-7.4	5.8-6.2	7.8-8.3	7.8-8.3
Alkalinity (as $CaCO_3$)	15-18 mg/L	2 mg/L	20 mg/L	14 mg/L
(Halogen + Sulfate)/ Alkalinity Ratio	0.5-0.8	2.5-4.5	0.5	0.5

The treatment is "low carbonate" corrosion control. On the Cedar, pilot tests have shown that lime treatment alone is the most effective and economical corrosion treatment for that supply.[8] On the Tolt, it will be accomplished with a combination of lime and soda ash.[9]

Materials Selection Program

The other half of the corrosion control plan includes a program relating to selection and use of corrosion resistant piping materials in home and building plumbing systems. The amendments listed below were made to the Seattle Plumbing Code[10] to either require or allow use of selected materials.

Plastic Water Pipe --

The SWD has used plastic pipe for underground potable water service connections for several years. The Seattle Plumbing Code[10] allowed plastics only for drains, wastes, and vents, but prohibited their use above ground, and inside of buildings for hot and cold water distribution. Working with the Seattle-King County Department of Public Health, the agency that enforces the plumbing code, and the Plumbing Code Advisory Committee (made up of representatives from the plumbers union, the mechanical contractors, architects, engineers and various public officials); the Seattle Plumbing Code has been amended to allow the use of both polybutylene (PB) and chlorinated polyvinyl chloride (CPVC) in homes and buildings.

Dielectric Insulator --

Direct connection of dissimilar metals can cause a corrosion rate increase in a localized area through galvanic action. To reduce the possibility of this occurrence, the Seattle Plumbing Code was amended to require the use of approved dielectric insulators when connecting piping made of dissimilar metals.

Copper and Galvanized Piping --

Based on the results of the pilot and field tests, copper piping will be significantly more resistant to corrosion than will galvanized piping. Further, thick-walled copper pipe will last longer than the thin-walled pipe. Consequently, it has now become the policy of the SWD to encourage use of thick-walled copper pipe (Types K and L) and discourage use of thin-walled Type M piping. Since galvanized pipe will continue to experience a relatively high rate of corrosion, even with chemical treatment, use of galvanized pipe in new construction is also discouraged. Legislative code changes to accomplish this end will be considered in the future.

Solder Materials --

The use of solder containing a high percentage of lead to join copper pipe creates a problem from a lead leaching standpoint. To reduce the potential for the entrance of lead into the water from this source, the maximum allowable lead in solder has been limited by amending the Seattle Plumbing Code. The amendment requires that solder have a lead content of no more than 0.20%. This amendment will require most contractors to use 95/5 tin/antimony solder.

Lead Pipe --

To reduce exposure of the customer to lead through ingestion of drinking water, lead pipe was removed from the Seattle Plumbing Code several years ago.

Asbestos Cement Pipe --

To reduce further additional contamination of the water supply from the leaching of asbestos fibers from asbestos cement (A/C) pipe, this pipe was removed from the approved list.

In summary, the materials selection program presented above is tailored to the water treatment program and will provide additional protection to the home and building piping in Seattle.

95/5 Tin/Antimony Solder

One of the few solders which will now conform to the Plumbing Code is 95/5 tin/antimony solder. 95/5 tin/antimony solder is made up of approximately 95% tin and 5% antimony; however, some impurities can be present. The ASTM standards for tin/antimony solder are presented in Table 3.[11]

PUBLIC HEALTH CONSIDERATIONS OF ANTIMONY[12]

The specifications of tin/antimony solder into the Plumbing Code raises a concern of additional human exposure to antimony. Humans are exposed to antimony from sources including ambient air, food, occupational exposure, and drinking water. Ambient concentrations of antimony in air sampled at various stations throughout the United States averaged 2×10^{-7} parts per million (ppm) with a 3.32×10^{-5} ppm maximum.[13] In an epidemiological study relating tap water constituents and cardiovascular disease, approximately 15% of the samples tested showed the presence of antimony with a maximum value of 3.5 micrograms per liter (µg/L), a minimum value of 0.5 µg/L and a mean value of 1.4 µg/L.[14] An average water intake of 2 liters (L) per day gives a daily intake of antimony from water of 2.8 µg.

The most significant source of human exposure to antimony is from diet. Murthy, et al (1971)[15] analyzed diet samples for antimony from 28 institutions in 28 different states. The foods tested contained an average of 0.361 mg of antimony per kg of food. An average food consumption of 1.90 kg per day gives a daily intake of antimony from food of 0.686 mg.

TABLE 3. ASTM % COMPOSITION OF 95/5 TIN/ANTIMONY SOLDER

Alloy Grade	Tin Desired	Lead Nominal	Antimony Min.	Antimony Desired	Antimony Max.	Bi Max.	Cu Max.	Fe Max.	Al Max.	An Max.	As Max.
95/5 Sn/Sb	95	0.20	4.5	5.0	5.5	0.15	0.08	0.04	.005	.005	0.05

Short-term, high level exposure to antimony has major physiological effects, especially on the heart, lungs, and kidneys, and on blood chemistry. The lowest reported lethal dose for humans is 100 mg with the lethality probably due to cardiotoxicity.[16]

Very little information, however, has been recorded on the biological effects of long-term, low level antimony exposure. Since the levels of antimony found in drinking water are low in comparison to the amounts required to cause acute toxicity, it is difficult to assess the public health significance of this exposure source.

STUDY OBJECTIVES

The formal objective of this research effort was to determine whether replacing lead-based solders with 95/5 tin/antimony solder in Seattle's Plumbing Code will ultimately result in increased antimony and reduced lead concentrations in tap water. The scope of work included:

1. Investigating the mechanisms and types of corrosion of 95/5 tin/antimony solder.

2. Predicting the metal leaching characteristics of tin/antimony solder.

3. Comparing lead leaching potential from tin/antimony and lead/tin solders exposed to control, lime/soda ash, lime/bicarbonate, and lime/bicarbonate/silicate treated Tolt water.

SCOPE OF STUDY

The study was divided into the following research phases:

Theoretical

The theoretical analysis included research into the corrosion characteristics and corrosion products of the solder via the construction of pe-pH diagrams (Pourbaix) and an examination of the various possible galvanic couples resulting from the use of the solder.

Laboratory

The laboratory tests entailed conducting short-term staic coupon tests. The coupons, which were constructed of tin/antimony solder and copper sheets, were placed in separate, sealed, water-filled containers and allowed to stand at 20°C for varying time periods. The amount of total and dissolved antimony leaching from the coupons was then measured.

Field

The field tests consisted of a sampling program at the University of Washington. The University since 1968 has been using tin/antimony solder

on all construction and remodeling of building plumbing systems. For the sampling program, overnight standing water samples were taken at several buildings which contained tin/antimony soldered plumbing. These samples were analyzed for metal content (lead, copper, zinc and antimony), pH, conductivity and alkalinity.

Metal Leaching

Side-by-side metal leaching tests were performed on tin/antimony and lead/tin soldered copper pipe which compared lead, copper, tin and antimony leaching. This is a new method which has been developed by the SWD as part of this study.

Scanning Electron Analysis and X-Ray Diffraction

An examination of the corrosion films formed on tin/antimony solder and lead/tin solder was conducted using scanning electron analysis and X-ray diffraction. Corrosion films were analyzed from pipe samples collected at the University of Washington and from the metal leaching test loops.

STUDY ELEMENTS

The study consisted of the following primary elements that included various secondary topics.

Corrosion Control Through Materials Selection

1. Metal leaching potential of tin/antimony solder.
2. Comparison of metal leaching from lead/tin and tin/antimony solder.

Methods of Analysis

1. Theoretical analysis of corrosion products.
2. Metal coupon analysis techniques.
3. Development of a metal leaching evaluation method.
4. Field sampling.
5. Scanning electron microscopy and X-ray diffraction analysis.

Corrosion Characterization

1. Metal leaching.
2. Corrosion film formation.

SECTION 2

CONCLUSIONS

COMPARISON OF TIN/ANTIMONY AND LEAD-BASED SOLDERS

Lead leaching is substantially reduced and the subsequent increase in the antimony concentration of the drinking water is minimal if tin/antimony solder is used in plumbing systems in place of lead-based solder. The low metal leaching levels from 95/5 tin/antimony solder are mainly due to the sacrificial corrosion of tin and the formation of a tin oxide passivation film that protects the surface of the solder from corrosion.

The theoretical and experimental results obtained in this study indicate, therefore, that 95/5 tin/antimony solder has excellent potential for replacing lead-based solders in private and commercial plumbing systems. The public health effects of antimony in drinking water, though not yet clearly known, appear to be much less hazardous than the effects of lead in drinking water.

THEORETICAL ANALYSIS

The theoretical investigation of antimony corrosion, tin corrosion, and galvanic coupling relationships indicates that the leaching of antimony from the solder may be inhibited by two mechanisms:

1. Sacrificial Corrosion. Although antimony alone is quite soluble, the galvanic coupling relationships among the three metals that make up a soldered joint (tin, antimony and copper) indicate that the corrosion of tin is theoretically more favorable than the corrosion of antimony. Tin, therefore, may act as a sacrificial anode.

2. Passivation. Tin can be passivated by tin oxide. This passivation film may also cover the entire surface of the solder, thereby inhibiting metal leaching from the solder.

LABORATORY EXPERIMENTS

The laboratory static coupon tests demonstrated that antimony dissolution from tin/antimony solder is substantially less than the theoretical or observed dissolution from pure antimony metal. The highest antimony concentration observed in the tin/antimony solder coupon test was 3.7 µg/L,

while 18000 and 22000 µg/L of dissolved antimony were observed in the pure antimony metal corrosion tests.

These results support the theoretical conclusions already put forward. In the short term, the inhibition of antimony leaching from tin/antimony solder is a result of the galvanic coupling relationship (namely, the sacrificial corrosion of tin).

FIELD SAMPLING

The field sampling showed that in the long term, antimony corrosion is inhibited by the passivation of tin in the solder. Two distinct films were visible on the tin/antimony soldered joints that were removed from the buildings at the University of Washington. In addition, a detectable antimony concentration was found in only one of the building water samples tested.

PILOT TEST

Side-by-side metal leaching tests indicated that lead leaching is substantially less from tin/antimony solder than from lead/tin solder. Some lead initially leaches from the tin/antimony solder. But after approximately 6 weeks of testing, lead leaching was reduced to below the detection limit. Antimony and tin leaching from the 95/5 tin/antimony solder was also consistently below the detection limit.

SECTION 3

RECOMMENDATIONS

SEATTLE WATER DEPARTMENT

The SWD should:

- Continue to implement its program of materials selection, especially in regard to eliminating the use of lead-based solders.
- Further investigate the potential for lead leaching from copper pipe.
- Determine the severity of lead leaching from old copper and lead-based soldered plumbing systems.

U.S. ENVIRONMENTAL PROTECTION AGENCY

The EPA should:

- Recommend against the use of lead-based solders in soft water.
- Develop a standard method for the determination of metal leaching from plumbing materials.
- Publish a state-of-the-art manual for corrosion control of drinking water.

SECTION 4

EQUIPMENT DESIGN AND OPERATION

LABORATORY TESTS

Metal leaching coupons were made by soldering a piece of 95/5 tin/antimony solder to a copper sheet (see Figure 2). The solder and copper sheets were cut such that each specimen analyzed had surface areas of 9.7 cm^2 of solder and 24.5 cm^2 of copper. The coupons were only soldered at the end to provide for uniform surface areas.

METAL LEACHING TESTS

Metal Leaching Test Loops

Short sections 25-51 cm of 0.635 cm pipe were attached to continuous flow test units for comparison of copper, lead, tin and antimony leaching under three alternative treatments. The leaching pipe sections were made of copper, copper plus 50/50 lead/tin solder, and copper plus 95/5 tin/antimony solder. Two velocities were used in establishing corrosion films in the metal leaching sections. Dissolved metals were determined after periodic 22 to 24 hours contact with test water in the laboratory.

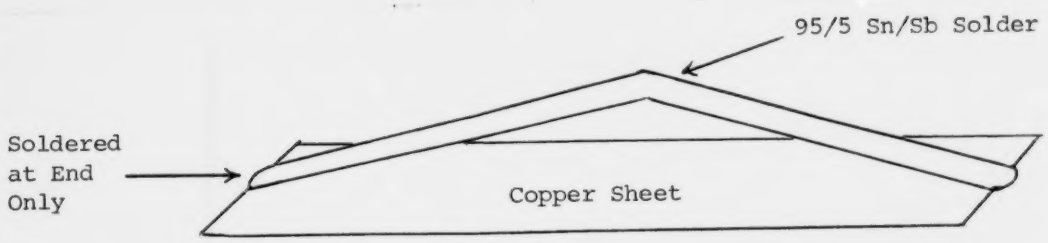

Figure 2. Tin/Antimony solder test specimen.

The construction of the 0.64 cm copper pipe with the solders can be seen in Figure 3. This design allows for uniform surface areas of copper and solder with intimate contact between the pipe and the solder. It exaggerates the metal leaching levels found in soldered copper pipe and therefore promotes meaningful comparison of the solders.

Small diameter pipes were chosen for the metal leaching tests because they have a high surface area to volume ratio which also exaggerates results; are easy to manipulate and control flow; and produce an adequate quantity of leachate for testing.

The metal leaching loops were attached to the Tolt River water pilot plant for testing. The pilot plant schematic is illustrated in Figure 4.

Chemical Treatment

Three treatment conditions were chosen to simulate average Tolt distribution system levels of pH and alkalinity. Carbon dioxide (CO_2) uptake in open distribution reservoirs and conversion of bicarbonate to CO_2 by post-reservoir chlorination will lower pH and alkalinity from treatment plant levels. Laboratory tests (presented in Volume II)[9] were performed to estimate these changes. Table 4 presents the expected average distribution water quality with lime/sodium carbonate, lime/bicarbonate, and lime/bicarbonate/silicate treatment.

Chemicals were fed by three AMF CUNO diaphragm pumps. The chemicals used were hydrated lime [$Ca(OH)_2$], sodium carbonate (Na_2CO_3), sodium bicarbonate ($NaHCO_3$), and N grade liquid sodium silicate (8.9% NaO/28.7% SiO_2). Feed solutions of the full strength chemicals [170 g $Ca(OH)_2$/151 L; 600 g Na_2CO_3/151 L; 4.2 kg $NaHCO_3$/151 L; 960 mL Na_2O-SiO_2/106 L] were mixed in plastic barrels. The feed solutions were then pumped from the solution barrels into the test units just before a static mixer.

SCANNING ELECTRON MICROSCOPY

Scanning electron microscopy analysis was performed by the Materials Analysis Center of the University of Washington's Department of Mining, Metallurgical and Ceramic Engineering.

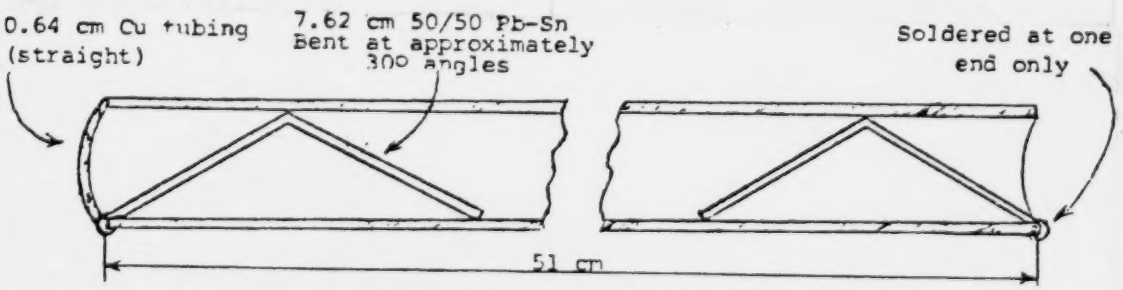

Figure 3. Copper pipe with 50/50 lead/tin solder.

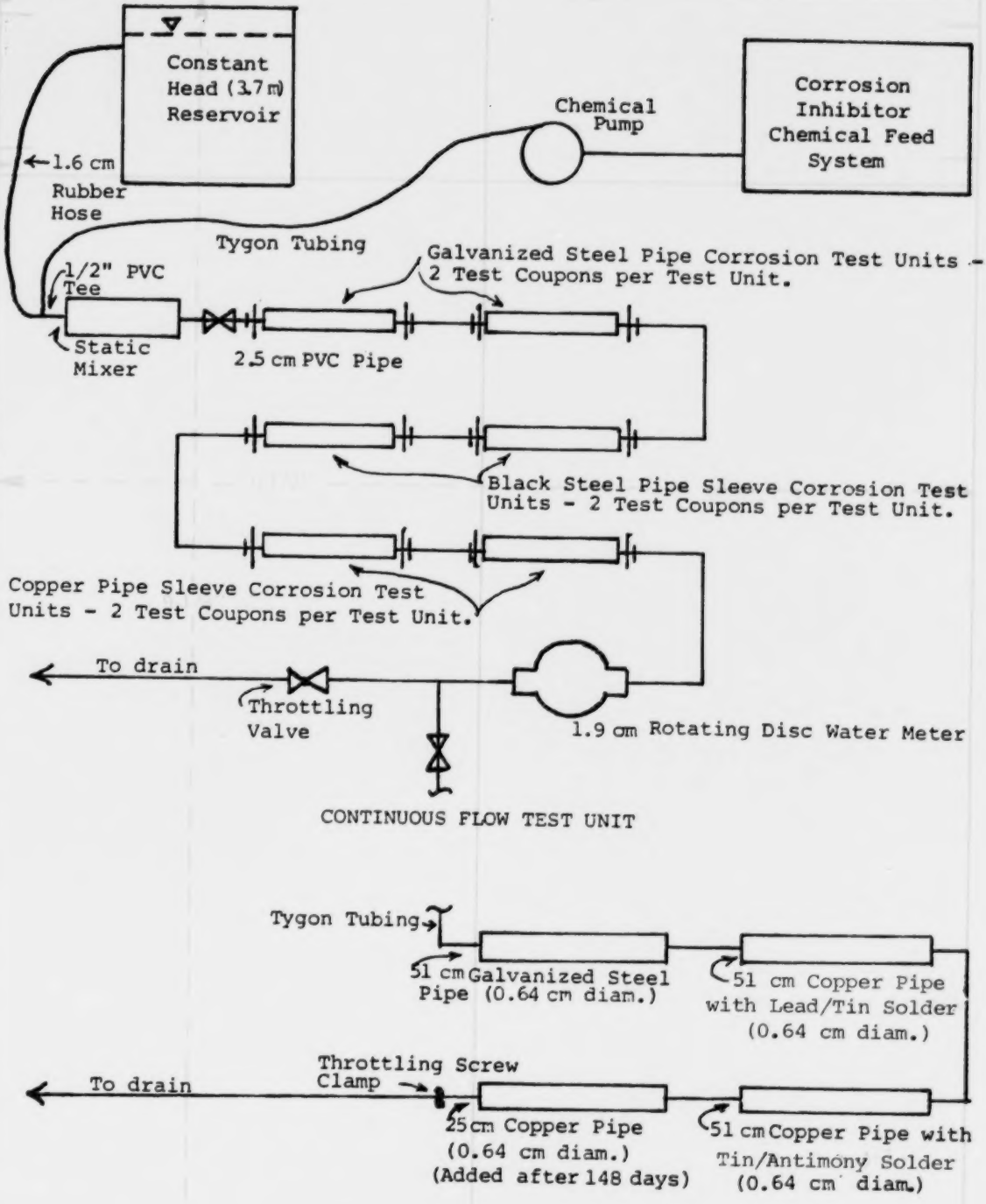

Figure 4: Pilot plant flow schematic.

TABLE 4. PILOT TEST CONDITIONS

Item	Loop #1 Control, Cedar Pipeline	Loop #2 Lime + Soda Ash	Loop #3 Lime + Bicarbonate	Loop #4 Lime + Bicarbonate + Silicate
pH	5.8-6.2	7.85	7.95	7.95
Alkalinity (mg/L $CaCO_3$)	2.0	14.5	21	21
Chlorine Residual (mg/L)	~.2-.4	~.2-.4	~.2-.4	~.2-.4
Lime Dosage, $Ca(OH)_2$ (mg/L)	0	1.5	4.5	4.5
Na_2CO_3 Dosage (mg/L)	0	10.0	0	0
$NaHCO_3$ Dosage (mg/L)	0	0	20.0	20.0
NaO/SiO_2 Dosage (mg/L as SiO_2)	0	0	0	4

The Center has a Cambridge Mark IIA Scanning Electron Microscope (SEM) capable of 200 Angstrom resolution, which was used in the secondary electron emission mode. The instrument has useful magnification to 50,000X and is unique in that is large depth of focus allows unusual observation and photography of three-dimensional structures not possible with other microscopic techniques.

The SEM is equipped with the EDAX Energy Dispersive X-Ray Detector for elemental analysis from fluorine to uranium in the atomic table. Qualitative analysis in the SEM mode was conducted with spectra generation typically less than a minute.

Mounting of the specimens and subsequent coating in a vacuum evaporator were performed by the Center. The specimens were coated with a 250 to 300 Angstrom layer of gold or palladium in order to provide a conductive surface.

SECTION 5

ANALYSIS AND PROCEDURES

THEORETICAL ANALYSIS[12]

In order to predict the electrochemical behavior and corrosion of the tin/antimony solder, a pe-pH diagram of the constituents of the solder was constructed. This diagram is primarily an aid for the interpreation of equilibrium constants, and provides an overview of the thermodynamically favorable reactions in a specific corroding system. Such a diagram, of course, only represents the information used in its construction.

Even though aqueous solutions do not contain free electrons, it is possible to define the relative electron activity as

$$pe = -\log (e) \tag{1}$$

The general redox half cell reaction where n electrons are transferred is

$$aRED \overset{K}{\rightleftharpoons} bOX + ne^- \tag{2}$$

where RED and OX represent the reduced and oxidized forms, respectively. This can be expressed as

$$pe = -\frac{1}{n} \log K + \frac{1}{n} \log \left[(OX)^b/(RED)^a\right] \tag{3}$$

The dimensionless parameter pe is related to the redox potential, E_H, by

$$pe = \frac{E_H(F)}{2.3\ RT} \tag{4}$$

where F is the Faraday constant, R is the gas constant and T is absolute temperature.

Substituting Eq. (4) into Eq. (3) yields the Peters-Nernst equation,

$$E_H = E_H^o + \left(\frac{2.3\ RT}{nF}\right) \log \left[(OX)^b/(RED)^a\right] \tag{5}$$

where

$$E_H^o = -\left[\frac{1}{n} \log K\right]\left[2.3\ RT\ F^{-1}\right] \quad (6)$$

Equilibrium constants can be calculated from free energy data by the equation,

$$\ln K = \frac{-\Delta G^o}{RT} \quad (7)$$

which at 25°C and converted to base 10 gives

$$\log K = \frac{-\Delta G^o}{1.364} \quad (8)$$

In Appendix A, values for the standard free energy of formation, ΔG^o, are listed for species involved in aqueous chemistry of antimony and tin. It should be noted that the precision of these values varies considerably, and that no attempt has been made to assess the probable errors associated with them. Pertinent equilibrium equations have been calculated for the antimony and tin corrosion processes using Eq. (1), (2), (3), and (8) and are presented in Appendix B. Several pe-pH diagrams have been constructed using these equations. The figures are drawn such that when the activity of dissolved metal is greater than 10^{-6} M (1 M for Figure 4), it is designated as being "dominant" compared to a solid species. The equilibrium activities of soluble species chosen represent common values found in Seattle's water. Sulfur, chlorine, and fluorine species are considered in Figures 6 through 9 with total activities of 10^{-5} M, 10^{-4} M and 10^{-4} M, respectively.

LABORATORY EXPERIMENTS

Laboratory experiments were conducted to verify the theoretical phase of this study as well as to provide practical information concerning the equilibrium concentration or maximum amount of antimony corrosion in a typical plumbing system.

Each specimen described in Section 4 was completely immersed in water samples collected from Seattle's North Beacon Reservoir and placed in a sealed 50 mL glass container. Samples were held under static conditions at 20°C, for periods of one-half to 98 hours.

In addition, 2 samples of antimony metal equalling the weight of antimony in the solder coupons, were tested as described above for a 70-hour period.

All samples were then analyzed for dissolved antimony and lead on an Instrumentation Laboratory, Inc., Atomic Absorption Spectrophotometer according to "Methods for Chemical Analysis of Water and Wastes," U.S. E.P.A. Technology Transfer, 1976.[17] When a furnace procedure was required, Model IL 555 controlled temperature furnace atomizer was used.

FIELD TESTS

The University of Washington has been using 95/5 tin/antimony solder on building plumbing systems since about 1968. The field study consisted of taking overnight standing 0.95 L tap water samples from 8 buildings on the University campus. The plumbing systems of the buildings tested range from 1 to 10 years in age and supposedly consist of copper piping bonded with 95/5 tin/antimony solder.

Running water samples (0.95 L) were taken at the mechanical room (where the building plumbing connects with the distribution system) of each building to obtain the characteristics of the inflow water. The standing water sample (0.95 L) was then taken from the potable water tap furthest from the mechanical room of the building.

The samples were then analyzed for temperature, conductivity, alkalinity, total and dissolved antimony, and total lead, copper and zinc, according to "Standard Methods"[18] and "Methods for Chemical Analysis of Water and Wastes."[17]

METAL LEACHING TESTS

The metal leaching test loop sections were periodically removed during the study. The pipe sections were filled with test water from their respective loops, the ends were sealed with parafilm and then allowed to stand horizontally at room temperature for 22 to 24 hours. After this period, the water was filtered through a 0.45 μm Millipore membrane filter, preserved (.5% HNO_3 in Pyrex) and analyzed for metals by flame and flameless atomic absorption spectrophometry (A.A.S.) according to "Methods for Chemical Analysis of Water and Wastes."[17] Background metal concentrations were determined after passing the test waters through the same filtration apparatus, and these values were subtracted from the levels found after leaching.

Table 5 lists the pipe types, metals analyzed and analysis techniques.

TABLE 5. PIPE TYPES, METALS ANALYZED AND ANALYSIS TECHNIQUES

pB, non-filterable	Flameless A.A.S.	Cu; Cu + 50/50 Pb/Sn solder. Cu + 95/5 Sn-Sb solder.
Cu, non-filterable	Flame A.A.S.	Cu; Cu + 50/50 Pb/Sn solder, Cu + 95/5 Sn/Sb solder.
Sb, non-filterable	Flameless A.A.S.	Cu + 95/5 Sn/Sb solder.

A.A.S. = Atomic Absorption Spectrophotometry.

QUALITY CONTROL OF METAL LEACHING TESTS

Daily pH, flow measurements and adjustments, and apparatus maintenance were performed to provide quality control over the test conditions. To document alkalinity, Na_2CO_3 feed in Loop #2, and $NaHCO_3$ feed in Loops #3 and #4, alkalinity tests were performed twice weekly. All measurements were performed according to "Standard Methods."[18] Complete water quality assurance data is summarized in Appendix C.

SECTION 6

RESULTS AND DISCUSSION

THEORETICAL ANALYSIS

Interpretation of pe-pH Equilibrium Diagram[12]

The domains of predominance of antimony and its compounds can be seen in Figures 5, 6 and 7, while the conditions for corrosion, passivation and immunity are found in Figure 8. In Figures 5, 6, 7 and 8, the stability domain of pure antimony has only a small area in common with the stability domain of water. Antimony is therefore a very slightly noble element which may be found in the natural state.

Sb(s) is the dominant antimony species in the aqueous environment described by Figure 8 for pH 4 below line A and for pH 10 below line B. At potentials above those corresponding to lines A and B in Figure 7, antimony is oxidized to soluble trivalent or pentavalent species.

As shown in Figures 7 and 8, the products of antimony corrosion reactions depend most strongly on pH and dissolved sulfur concentration. Active antimony corrosion may result in the formation of the trivalent forms, thioantimonate, SbS_2^-, and antimonyl cation SbO^+, or, at stronger oxidation potentials, the formation of pentavalent antimonates SbO_3^- [or $Sb(OH)_6^-$].

As shown in Figure 8, passivation by antimonous sulfide, Sb_2S_3, may occur at low pH values. No such protection is offered by antimony oxide coatings, since all the oxides (Sb_2O_3, Sb_2O_4, Sb_2O_5) are much too soluble to form a passivating film. Compounds of antimony and the halogens ($SbCl_3$, SbF_3, $SbBr_3$, SbI_3) are also too soluble to result in passivation [see Appendix B, Eq. (25) and (26)].

At high total dissolved antimony activity, $(Sb)_T$ = 1 M; however, both the trivalent antimony trioxide Sb_2O_3(s) and the pentavalent pentoxide Sb_2O_5(s) have areas of stability (Figure 5). Sb_2O_3(s) is thermodynamically stable in aqueous solutions at moderate oxidation potentials. Oxidizing agents such as nitric acid, chlorine, bromine, iodine and iodates readily oxidize Sb_2O_3(s) to Sb(V). Sb_2O_5(s) is thermodynamically stable at high redox potentials and in the presence of oxygen.

Another solid, the tetravalent antimony tetroxide Sb_2O_4(s) is thermodynamically unstable and decomposes into a mixture of Sb_2O_5(s) and Sb_2O_3(s). Konopik and Zwiaver (1952)[19] have shown that Sb_2O_4(s) in aqueous solutions behaves as a mixture of Sb_2O_5(s) and Sb_2O_3(s).

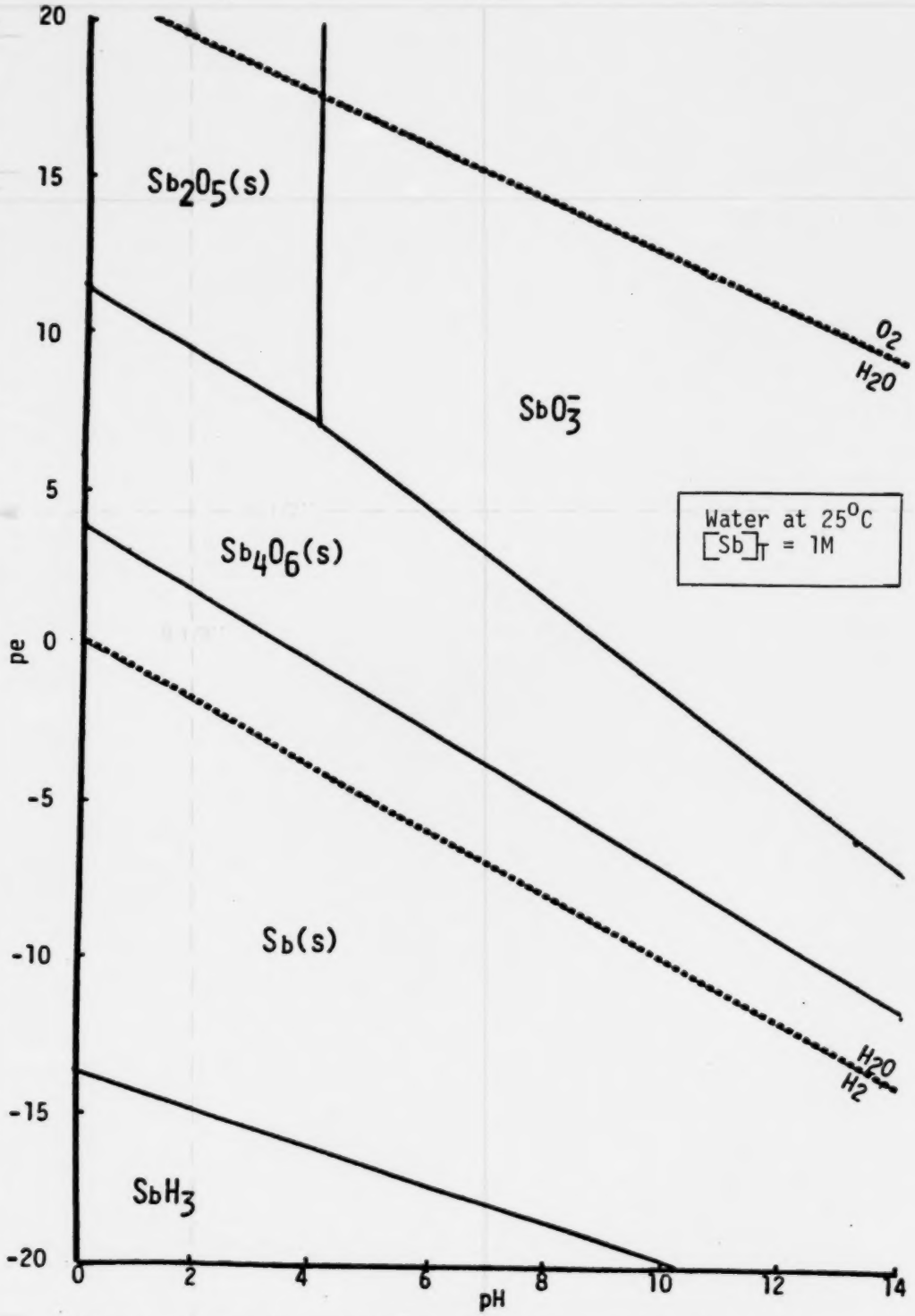

Figure 5. pe-pH equilibrium diagram for the system antimony

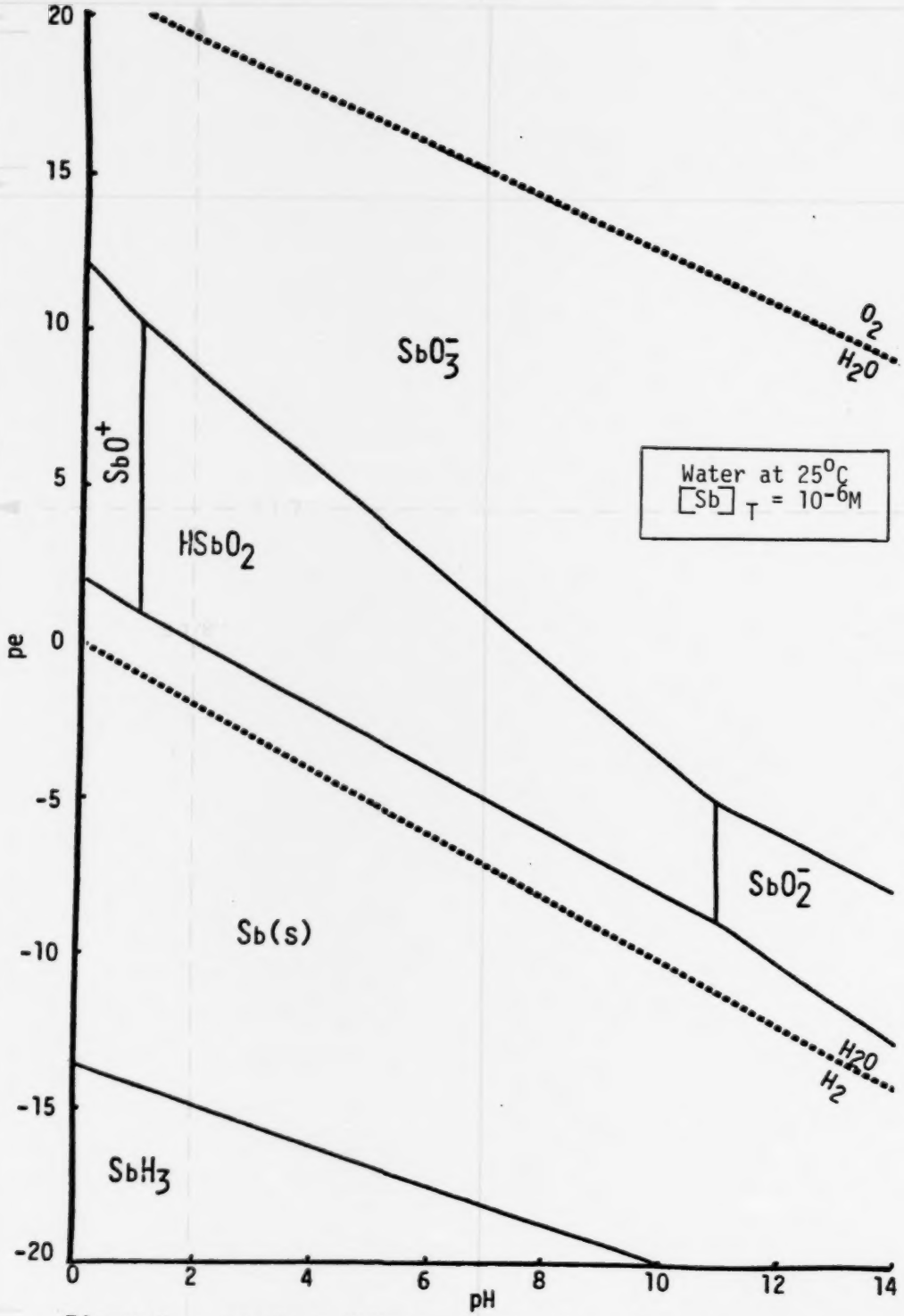

Figure 6 . pe-pH equilibrium diagram for the system antimony

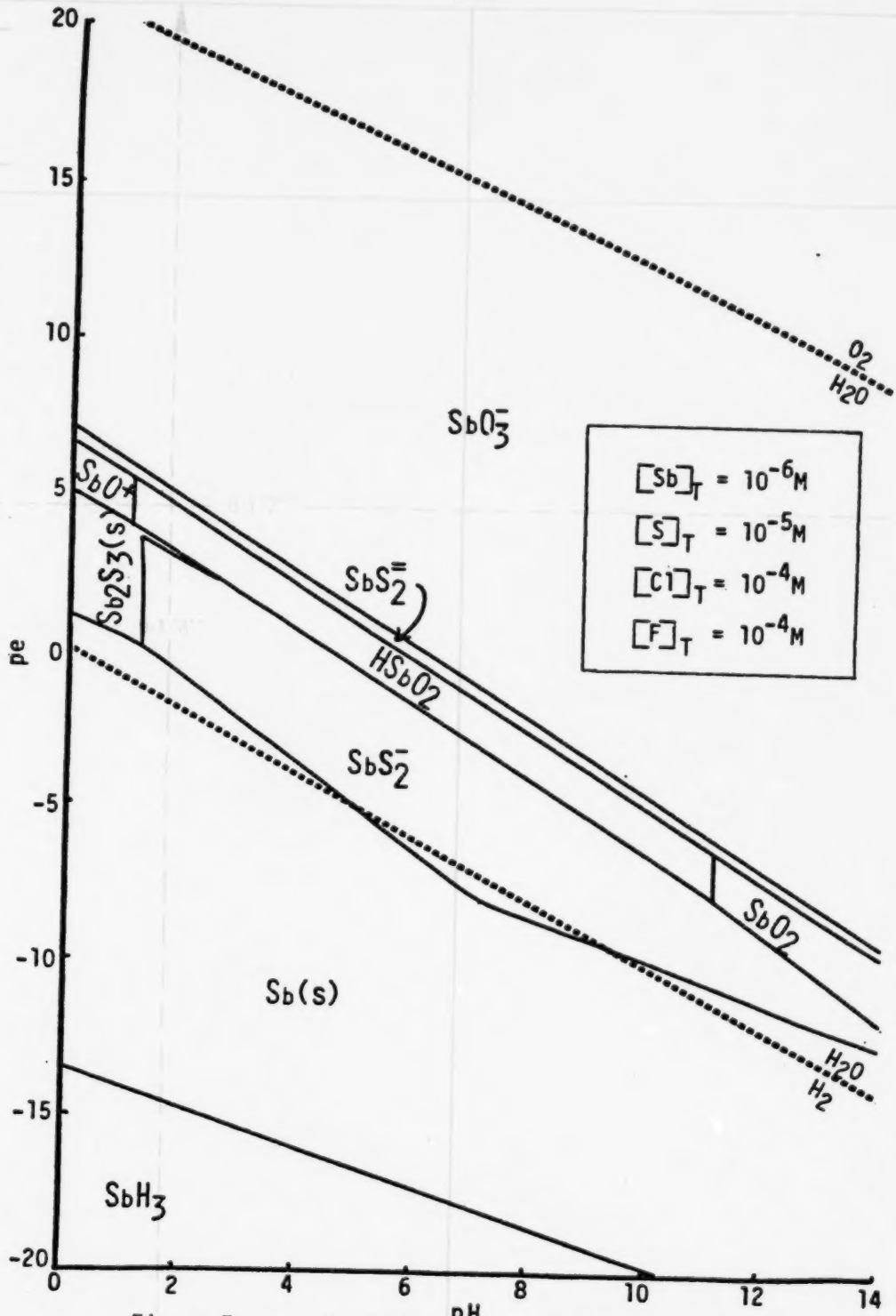

Figure 7. pe-pH equilibrium diagram for antimony.

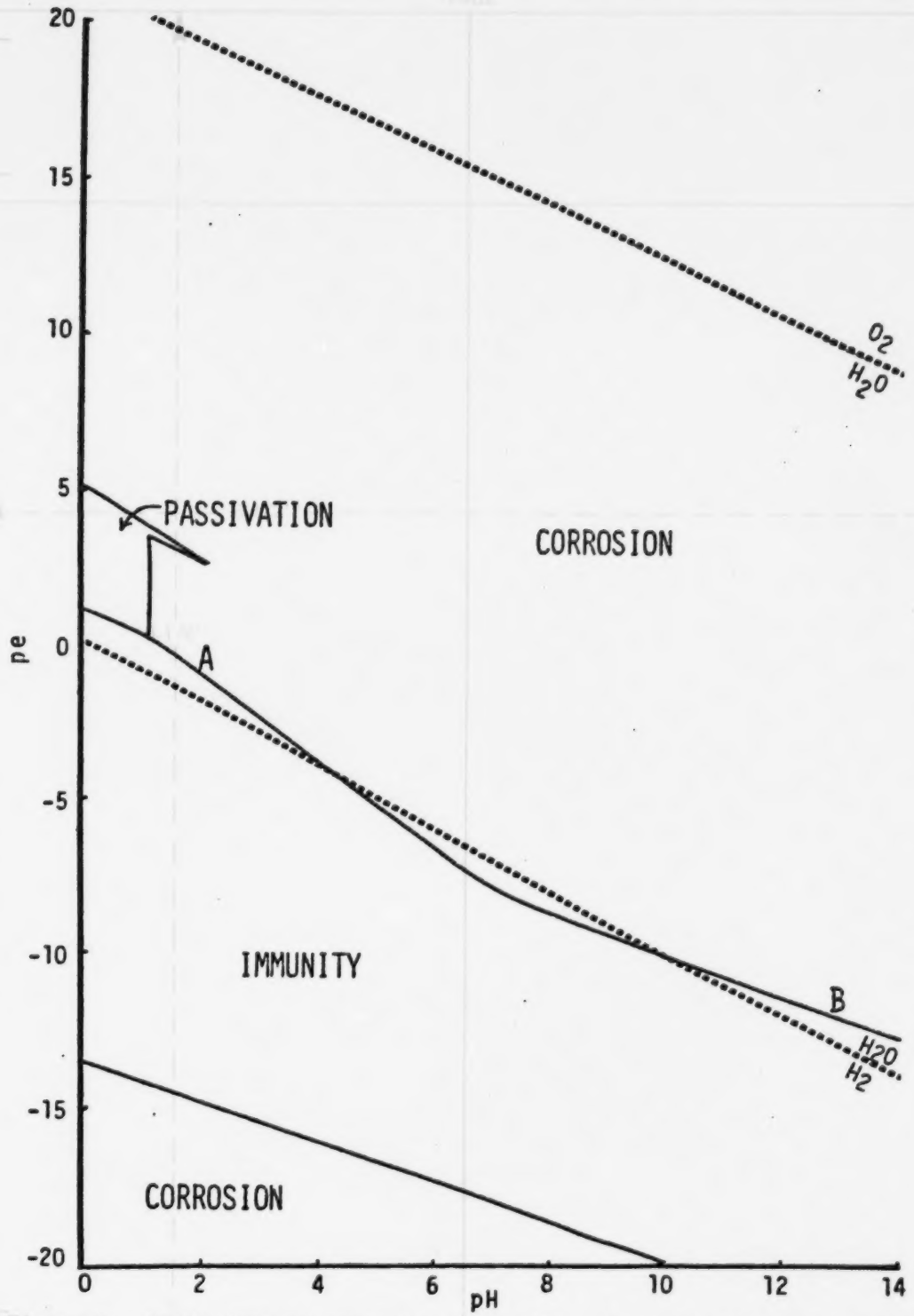

Figure 8. Conditions of corrosion, immunity and passivation of antimony.

As noted earlier, at lower total dissolved antimony concentrations, the antimony oxides are all soluble (Figure 6). $Sb_2O_3(s)$ dissolves readily to $HSbO_2^o$. $Sb_2O_5(s)$ forms antimonate SbO_3^-, and $Sb_2O_4(s)$ decomposes as stated above.

When antimony is the only metal exposed to a corrosive solution, the resultant oxidation products would be those discussed above. However, when antimony alloyed with other metals, such as in tin/antimony solder, is immersed in a corrosive solution, the degree of antimony corrosion may be much greater or much less than if only antimony were exposed to the solution.

Since tin is the major constituent in the solder, an inquiry is needed into the corrosion of tin and the effects of tin corrosion on antimony. Pourbaix (1966)[15] states that metallic tin can be passivated by tin oxide, SnO_2. In Figure 9, the oxidation products for the tin-water system are shown. Figure 10 depicts the theoretical conditions for corrosion, passivation and immunity for tin. It follows from Figure 10 that, theoretically, tin will resist corrosion in slightly acid, neutral and moderately alkaline solutions. Tin can, however, form soluble chloride, fluoride and sulfide complexes. Furthermore, Pourbaix reports that these theoretical conclusions are in fairly good agreement with the observed data.[20]

Galvanic Corrosion

Galvanic corrosion can be defined as the increased corrosion resulting from the electrical contact or coupling of two metals. Such a situation may exist during the corrosion of tin/antimony solder. However, in this case there are three metals to be considered. Two of the metals, tin and antimony, are major constituents of the solder; the third, copper, must also be considered since it is in direct electrical contact with the solder.

Therefore, there are three galvanic couples which may exist in this system: copper/antimony, copper/tin and tin/antimony. Although both metals in a couple may corrode, the metal with the lower redox potential will generally corrode at a greater rate while the metal with the greater potential will provide cathodic sites. The corresponding redox potentials for antimony, copper and tin are presented graphically in Figure 11. The data in Figure 11 indicate that copper/tin will form the strongest galvanic couple.

In the copper/antimony couple, the anode will be the antimony and the cathodic sites will be on the copper. Cathodic reactions at the copper water interface may include the reduction of dissolved oxygen,

$$2e^- + 2H^+ + 1/2\ O_2 \rightarrow H_2O; \qquad (9)$$

the reduction of the hydrogen ion,

$$2e^- + 2H^+ \rightarrow H_2; \qquad (10)$$

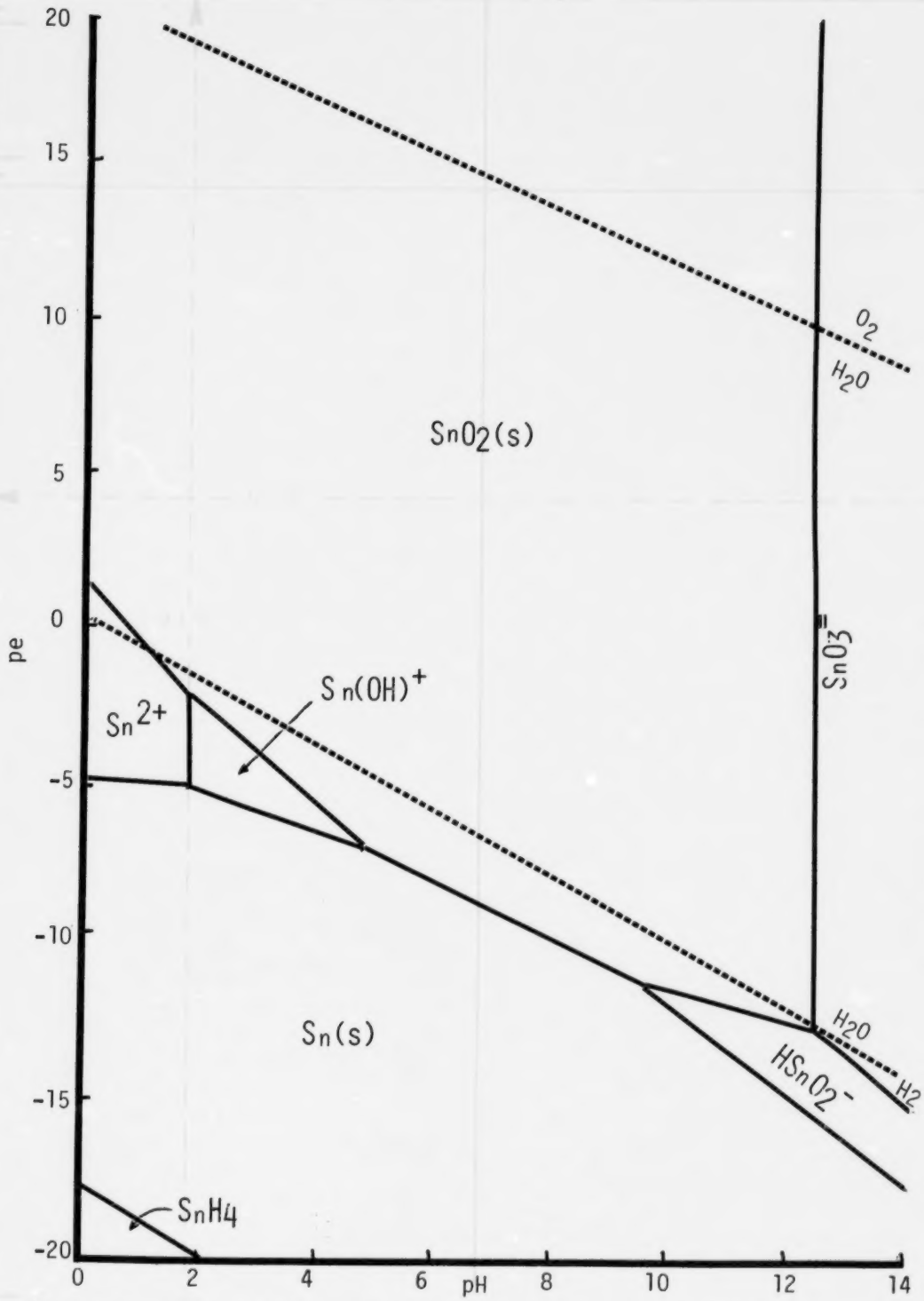

Figure 9. pe-pH equilibrium diagram for tin at 25°C and $[Sn]_T = 10^{-6}M$.

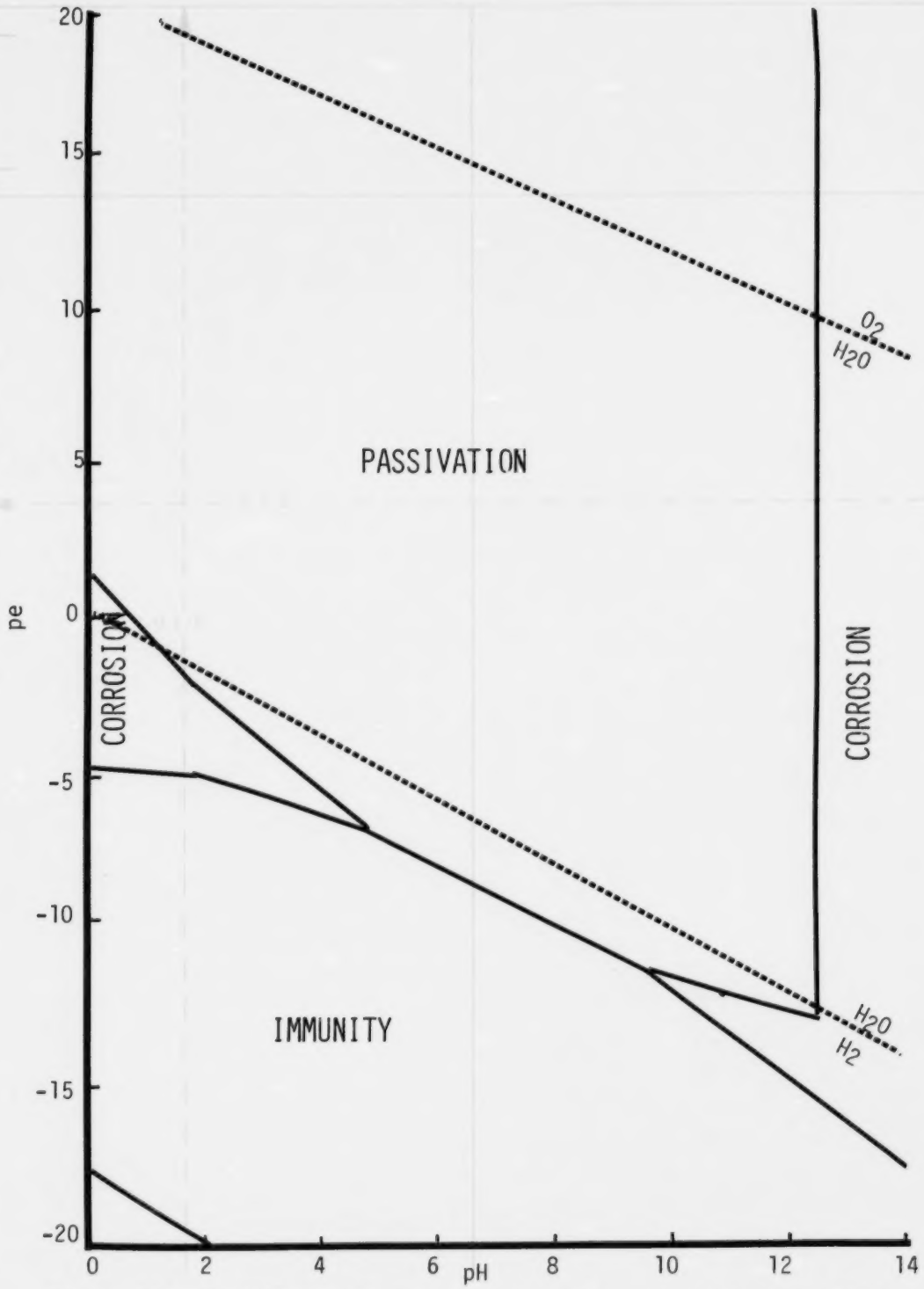

Figure 10. Conditions of corrosion, immunity and passivation of tin.

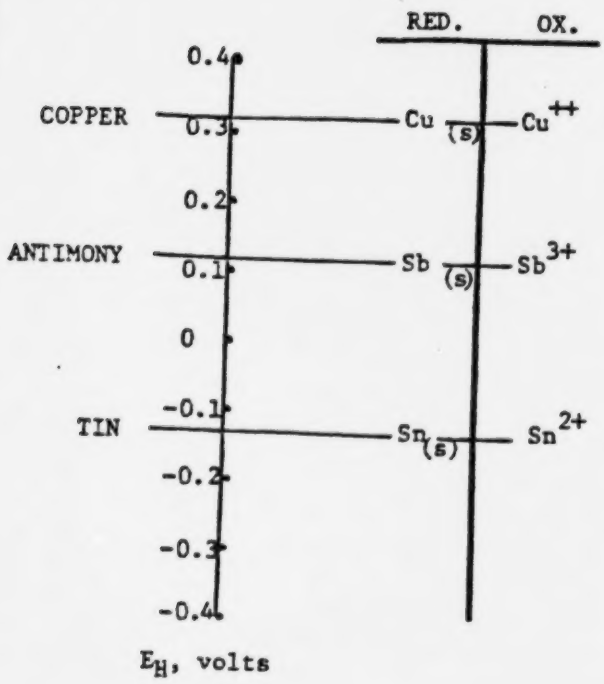

Figure 11. Selected standard equilibrium potentials.

or the plating out of copper ions as copper metal,

$$Cu^{2+} + 2e^{-} \rightarrow Cu(s) \quad (11)$$

Therefore, the overall reaction in the copper/antimony couple may be any of the following:

$$4Sb(s) + 12H^{+} + 3O_2 \rightarrow 4Sb^{3+} + 6H_2O \quad (12)$$

$$2Sb(s) + 6H^{+} \rightarrow 2Sb^{3+} + 3H_2 \quad (13)$$

$$Sb(s) + 3Cu^{2+} \rightarrow 2Sb^{3+} + 3Cu(s) \quad (14)$$

Tin will be the anode in the copper/tin galvanic couple with the possible cathodic reactions (9), (10) and (11) occurring at the cathodic site provided by the copper. The reactions for the copper/tin couple are:

$$Sn(s) + 2H^{+} + 1/2\ O_2 \rightarrow Sn^{2+} + H_2O \quad (15)$$

$$Sn(s) + 2H^{+} \rightarrow Sn^{2+} + H_2 \quad (16)$$

$$Sn(s) + Cu^{2+} \rightarrow Sn^{2} + Cu(s) \quad (17)$$

In the antimony/tin couple, tin is the anode and antimony the cathode site. The possible cathodic reactions are again (9) and (10), as well as the plating out of antimony ions as metal antimony,

$$Sb^{3+} + 3e^{-} \rightarrow Sb(s) \quad (18)$$

The corrosion reactions for the antimony/tin couple are therefore Eq. (15) and (16) and

$$3Sn(s) + 2Sb^{3+} \rightarrow 3Sn^{2+} + 2Sb(s) \quad (19)$$

Summary of Theoretical Analysis

The theoretical analysis of antimony corrosion, tin corrosion and the galvanic coupling relationships indicate that metal leaching from the solder may be inhibited by both the sacrificial corrosion of tin in the copper/tin galvanic couple and the formation of a protective tin oxide passivation film.

LABORATORY EXPERIMENTS

The coupon tests failed to show a strong correlation between antimony leaching and time, although a general increase in antimony concentration with time was observed (Table 6). Since each coupon was prepared separately, this variation in the reaction rates and antimony concentrations may be partly due to inter-sample differences.

TABLE 6. COUPON TEST RESULTS

Time (Hours)	pH	Antimony µg/l	Lead µg/l
Tin/Antimony Soldered Copper Coupons			
0	7.03	< 0.6	NA
0.5	7.00	< 0.6	NA
0.5	7.08	0.6	NA
1.0	7.08	< 0.6	NA
2.0	7.10	< 0.6	NA
2.0	7.09	1.9	NA
3.0	7.14	1.3	NA
3.0	7.17	0.6	NA
5.0	7.16	0.8	NA
5.0	7.16	2.0	NA
7.0	7.10	< 0.6	NA
9.0	7.11	< 0.6	12.0
9.0	7.12	1.0	22.0
27.	7.00	1.5	23.0
27	6.82	3.7	13.5
49	7.00	1.2	NA
49	7.04	2.0	NA
98	7.00	3.2	NA
98	7.00	2.0	NA
Pure Antimony Metal			
70	7.10	18,000	NA
70	7.10	22,000	NA

NA = Not analyzed.

The coupon tests, however, did demonstrate that the antimony concentrations in the water in contact with pure antimony metal were substantially greater than those in contact with the tin/antimony soldered coupons. The greatest antimony concentration leached in tin/antimony soldered coupon was 3.7 µg/L, while 18000 µg/L and 22000 µg/L were leached in the pure antimony corrosion test. This implies that antimony corrosion may be inhibited by the sacrificial corrosion of tin in the short term.

All four of the samples tested for lead contained concentrations between 12.0 and 23.0 µg/L. This lead presumably comes from impurities in the tin/antimony solder.

A white powder-like compound precipitated from solution and adhered to areas on the solder during the static tests. This white compound is most likely the passivation layer of SnO_2 as predicted in the theoretical corrosion diagram for tin (Figure 8). Beneath these white areas, black stains appeared on the surface of the solder. Pourbaix (1966)[19] reports that black stains on tin are a protective layer made up of a mixture of SnO(s) and SnO_2(s). Brennert (1935)[21] was able to isolate this layer and determined its composition to be approximately 80% SnO(s) and 20% SnO_2. The black color of the stains would therefore be due to the presence of black SnO(s). The presence of these oxides indicates that in the long term, the dissolution of antimony from the solder may also be inhibited by the formation of a passivating film.

FIELD SAMPLING

The results of the field sampling at the University of Washington are presented in Table 7. In only one of the buildings sampled, the Oceanography Teaching Building, did the overnight-standing tap water contain detectable antimony (0.6 µg/L).

Three soldered joints were removed from the plumbing in the Engineering Library and examined visually. The surface of the solder in each of the joints appeared to be completely covered by a white film. As in the coupon tests, a balck film (SnO) was under the white film (SnO_2). The presence of these films, when considered with the antimony levels observed in the field tests, strongly indicate that in the long term, antimony leaching from the solder is inhibited by passivation. These corrosion films were also analyzed by SEM and X-ray diffraction. Photographs from the SEM and EDAX tests are presented in Appendix D.

Temperature measurements were taken to give a rough indicator of the "age" of the standing water. The temperature difference between the standing water and the inflow water (mechanical room sample) qualitatively indicates lengthy standing times. Six out of the 8 building water samples appeared to have lengthy standing times.

Conductivity and alkalinity were measured to determine the source of the water (Cedar or Tolt watershed) and clearly indicate the Cedar as the source. Typical Cedar and Tolt water quality values are shown in Appendix E.

TABLE 7. TAP WATER SAMPLES FROM UNIVERSITY OF WASHINGTON BUILDINGS

Building	Year Occupied	Sample	Temp °C	Conductivity μmhos/cm	Alkalinity mg/l $CaCO_3$	Antimony μg/l Total	Antimony μg/l Dissolved	Lead μg/l	Copper μg/l	Zinc μg/l
Oceanography Teaching	1969	Mechanical Room	21.0	56.7	18.6	< 0.6	< 0.6	4.5	25	10
		Standing	25.0	55.2	18.6	2.0	1.0	2.0	2500	19
South Campus Center	1969	Mechanical Room	18.0	61.6	18.8	< 0.6	< 0.6	3.5	30	15
		Standing	26.0	59.4	17.6	< 0.6	< 0.5	3.0	470	40
Health Sciences Wing T	1973	Mechanical Room	15.0	60.5	19.4	< 0.6	< 0.6	< 1.0	< 10	15
		Standing	22.5	51.4	15.4	< 0.6	< 0.6	1.0	1600	210
Condon	1974	Mechanical Room	32.0	58.9	17.4	< 0.6	< 0.6	8.5	20	360
		Standing	23.5	61.0	18.8	< 0.6	< 0.6	11.5	730	120
Waterfront Activities	1977	Mechanical Room	18.0	62.5	20.0	< 0.6	< 0.6	2.0	100	280
		Standing	27.5	58.9	20.0	< 0.6	< 0.6	11.0	250	120
Hall Health Center	1978	Mechanical Room	23.0	70.3	26.0	< 0.6	< 0.6	1.5	10	240
		Standing	23.0	55.1	19.8	< 0.6	< 0.6	3.0	690	70
Loew	1979	Mechanical Room	20.5	59.0	17.8	< 0.6	< 0.6	4.5	35	35
		Standing	21.0	60.5	20.2	< 0.6	< 0.6	1.0	770	25
Engineering Library	1969	Mechanical Room	20.5	59.0	17.8	< 0.6	< 0.6	4.5	35	35
		Standing	22.5	62.6	20.0	< 0.6	< 0.6	1.5	750	40

To verify that no lead solders, lead pipes, or galvanized pipes were employed in each building's plumbing system, analyses for lead and zinc were conducted. Since zinc concentrations showed no appreciable increase in the standing water samples relative to the mechanical room samples, galvanized piping was apparently not used in any of the plumbing systems.

The lead analyses indicate that lead pipe and/or lead solder were not used in 7 of the 8 buildings sampled. The Waterfront Activities Building was the only building in which there was a significant increase in the lead concentration of the standing water sample relative to the concentration of the mechanical room sample.* The low levels of lead found in these samples also indicate that lead concentrations observed in the coupon tests are not maintained over long periods. There is an initial leaching of the small amount of lead contained in the solder, but as the solder joint ages, lead leaching in minimized.

All of the buildings tested showed substantially greater copper concentrations in the standing water samples than in the mechanical room samples. Values exceeding the proposed Federal secondary standards for copper (1 mg/L) were found in the Oceanography Teaching Building (2.50 mg/L) and in the Health Sciences Building (1.70 mg/L). This indicates that a substantial degree of copper corrosion occurs in the building plumbing system.

METAL LEACHING

General

Untreated and treated Tolt water's impacts on the leaching of copper, lead, antimony and tin during 24-hour contact time with tin/antimony and lead/tin soldered copper pipe was determined as described in Section 5. The tests were conducted at an initial velocity of 0.12 m/s. At 140 days, the velocities were increased for the remainder of the test. The velocity increase was designed to increase metal leaching; however, this did not occur in all cases.

Copper Pipe

Copper pipe was analyzed for copper and lead leaching. Figure 12 demonstrates a reduction in copper leaching with treatment and also shows a general increase in copper leaching with time. Surprisingly, lead was found to leach from copper pipe. This is most likely due to lead impurities that may be present in copper pipe.

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*Welders often overlap the 95/5 Sn/Sb solder with 50/50 Pb/Sn solder to prevent gapping. Also, some contractors or welders may "cheat" and use the 50/50 Pb/Sn in place of the 95/5 Sn/Sb because of its lower price and easier workability. This most likely accounts for the high lead values observed. (Personal communication, E. Weber, Physical Plant Foreman, University of Washington.)

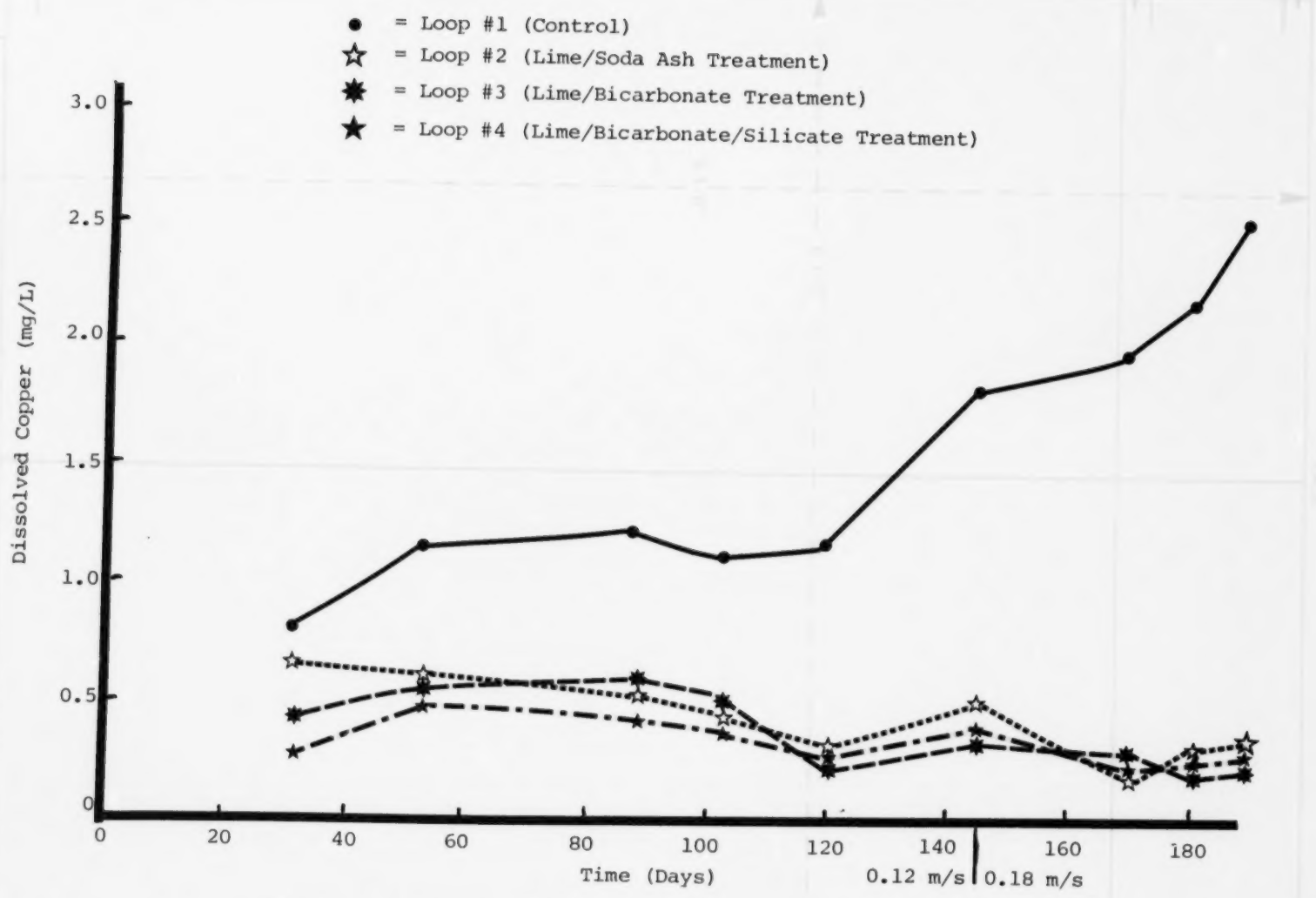

Figure 12. Copper leaching from copper piping in Tolt water.

A section of unused 1/4" tubing, utilized to construct the test units, was dissolved in nitric acid and analyzed for lead by flame A.A.S. with background correction to determine the source of lead. In this one test, lead was found to make up .005% of the copper pipe.

ASTM alloy specifications (B 42-80) for copper pipes make no specification for lead content, only copper and phosphorus content. In copper used for other applications, as much as .004% lead is permitted. Copper pipe manufacturers have indicated that copper water tubing is commonly manufactured from recycled copper products and this practice may result in the introduction of lead impurities to the pipe. This suggests that a survey of lead in various brands and lots of copper piping may be necessary to determine whether this is a significant source of lead in drinking water.

Copper Pipe and 50/50 Lead/Tin Solder

Copper and lead leaching from copper pipe with 50/50 lead/tin solder was analyzed and is presented in Figures 13 and 14. Tin leaching was also measured but was below the detection limit of 100 µg/L.

Again, copper leaching was reduced with treatment and generally increased with time in the control loop. Dissolved copper levels were usually lower in the copper plus 50/50 lead/tin leaching loop than in the copper pipe test loop. This is a result of the sacrificial corrosion of the solder when placed in contact with the more noble copper.

Dissolved lead levels generally were initially quite high and decreased with time. Lead levels were lower in the treated water loops than in the control loop for the first part of the tests, but after approximately 120 days, the control loop showed lower lead levels than the treatment loop. This phenomena is due to the depletion of lead near the surface of the solder and the subsequent formation of a tin oxide film which protects the solder from further corrosion. This phenomena is discussed in detail later in this Section and in Volume II of the Seattle Distribution System Corrosion Control Study.[9]

The high lead levels which occurred at 87 days were the result of treatment breakdown. Approximately 6 to 12 hours before the beginning of the metal leaching tests, the chemical feed pumps failed, resulting in the treatment loops exposure to untreated water. This apparently caused a disruption in the protective film with high lead leaching as a result.

Copper Pipe and 95/5 Tin/Antimony Solder

Copper and lead leaching from copper pipe plus 95/5 tin/antimony solder was analyzed and is presented in Figure 15 and Table 8. Dissolved tin and antimony were also measured but both were under the detection limit of 100 µg/L and 1.4 µg/L, respectively.

The lack of a detectable antimony concentration in the leachate is quite significant because it supports the theoretical analysis already put forward and is in agreement with the field sampling results. From these

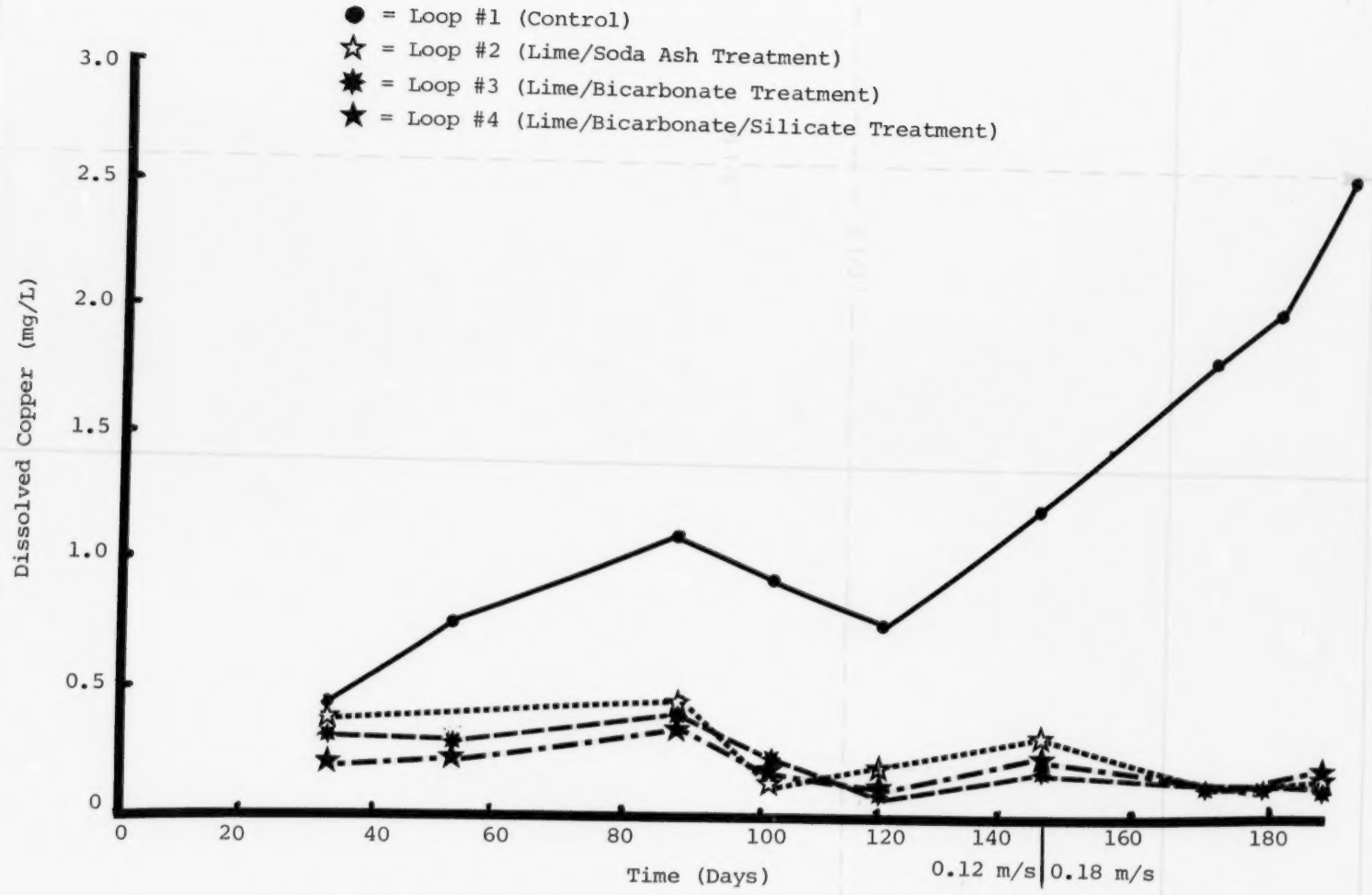

Figure 13. Copper leaching from copper piping and 50/50 lead/tin solder in Tolt water.

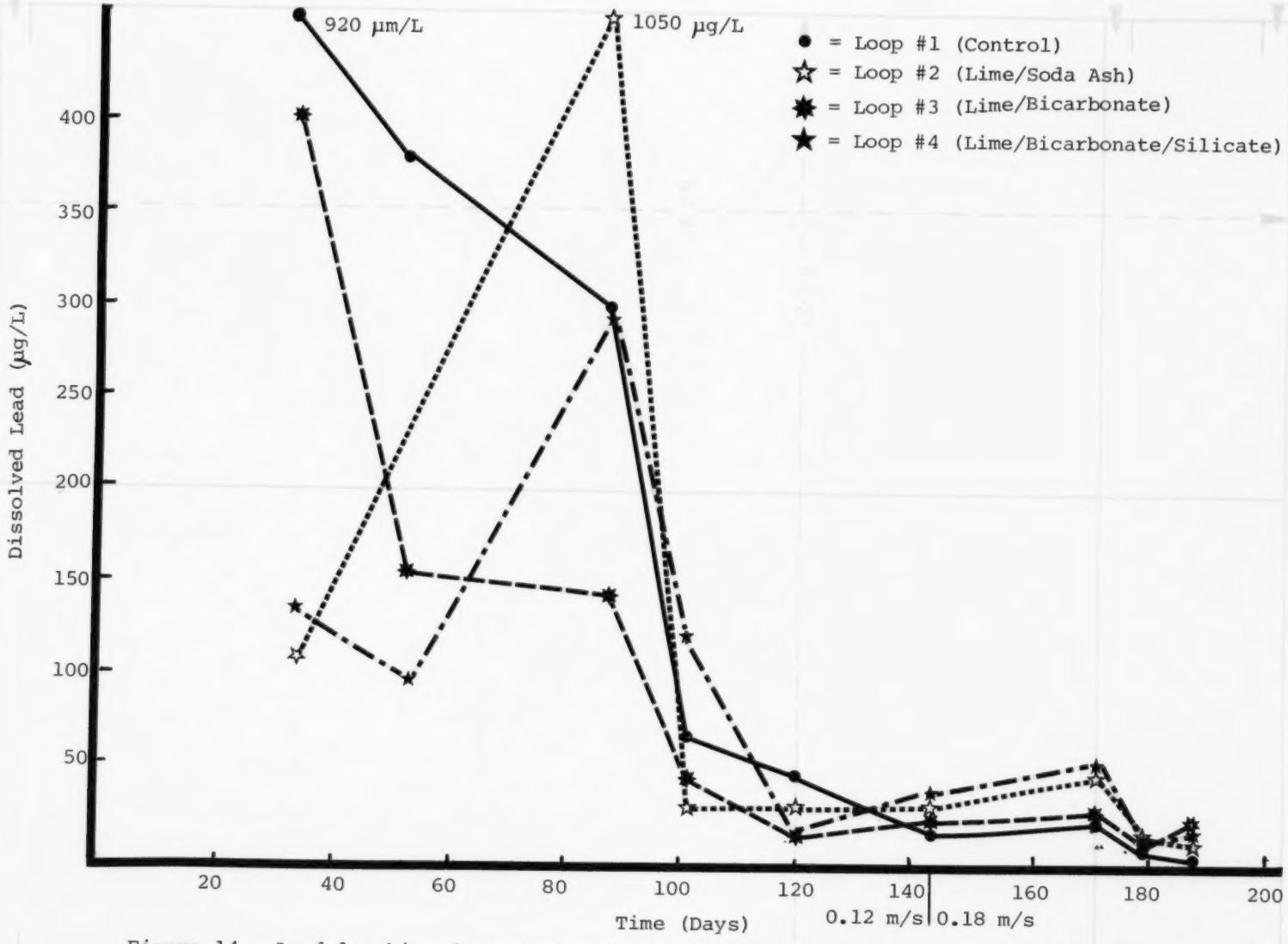

Figure 14. Lead leaching from copper piping and 50/50 lead/tin solder in Tolt water.

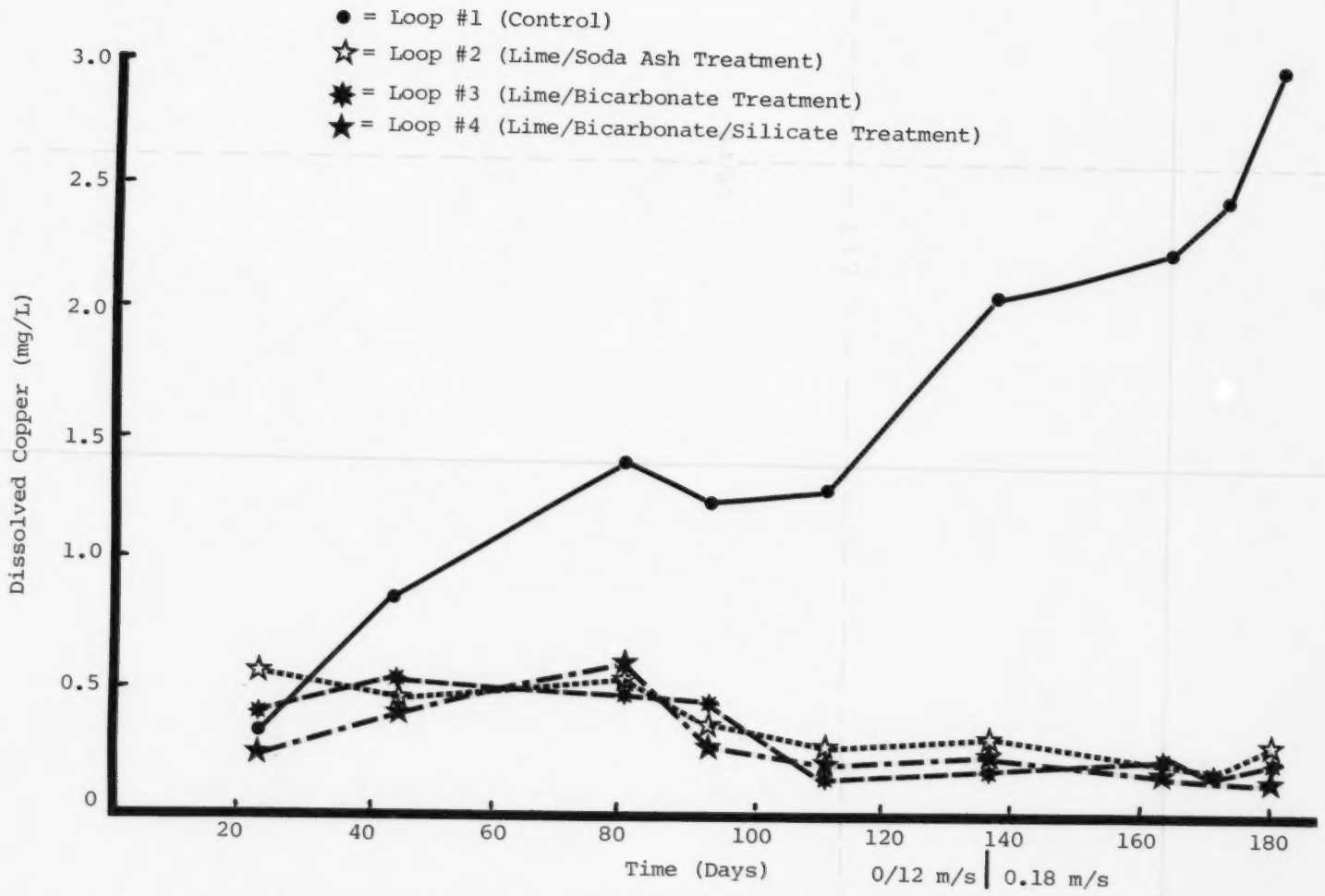

Figure 15. Copper leaching from copper piping and 95/5 tin/antimony solder in Tolt water.

TABLE 8. LEAD LEACHING FROM COPPER PIPE WITH 95/5 TIN/ANTIMONY SOLDER IN TOLT WATER

	Lead, µg/L			
Exposure (days)	Control	Lime/ Sodium Carbonate Treatment	Lime/ Bicarbonate Treatment	Lime/ Bicarbonate/ Silicate Treatment
26	113	65	4	1.5
45	17	3	11	<1
79	<1	35	<1	<1
93	1.5	2.5	<1	<1
112	2	<1	<1	<1
137	<1	<1	<1	<1
163	<1	NA	<1	<1
172	<1	<1	<1	<1
180	1½	<1	<1	<1

NA = Not analyzed.

results, it can be concluded that the antimony concentration in drinking water will not be substantially increased when 95/5 tin/antimony solder is used.

Figure 15 illustrates that, as in the previously mentioned metal leaching tests, copper leaching was reduced with treatment and generally increased with time. Copper leaching was initially lower in the copper pipe than in the copper plus 95/5 tin/antimony solder because of the sacrificial corrosion of the solder. With time, however, copper leaching became approximately equal in the two leaching sections. This is due to the formation of a passivation film on the solder which inhibits sacrifical corrosion.

Lead was found to initially leach from the tin/antimony solder, as demonstrated in Table 8. However, after approximately 74 days, lead levels were near or below the detection limit. Table 8 also shows that treatment significantly reduced lead leaching from the solder during the initial lead leaching period. Also, a spike occurred on Loop #2 when there was a breakdown in treatment. This is probably due to the deterioration of the corrosion film during changing water qualities.

Comparison of Metal Leaching from 95/5 Tin/Antimony and 50/50 Lead/Tin Solders

Figures 16, 17, 18 and 19 present lead leaching versus time from 95/5 tin/antimony solder and 50/50 lead/tin for the control loop and each treatment alternative. These figures clearly illustrate that a substantial reduction in dissolved lead concentrations is realized by replacing 50/50 lead/tin solder with 95/5 tin/antimony solder. This is especially true for the first part of the metal leaching tests where lead leaching from the lead/tin solder is very high. As the tests progressed, however, lead leaching from the lead/tin solder decreased to levels nearly as low as the tin/antimony solder leaching levels. The mechanisms for this reduction are discussed in the following Section and in the Volume II report. Although this decrease in lead leaching with time from the lead/tin solder may be a characteristic of premise plumbing metal leaching, it should be noted that the metal leaching test greatly accelerates corrosion and therefore the time scale used cannot be interpreted to be that of actual premise plumbing corrosion.

SCANNING ELECTRON MICROSCOPY

General

SEM and EDAX analyses were conducted on specimens from the laboratory tests, buildings from the University of Washington, and both the tin/antimony and lead/tin solder metal leaching loops. SEM and EDAX photographs are presented in Appendix D. To provide a conductive surface on the specimens, the laboratory test specimens and the specimens from the University of Washington were coated with gold (Au) and the metal leaching loop specimens were coated with palladium. It was found that the palladium coating was better suited to this EDAX analysis because the gold M lines were difficult to distinguish from the lead M lines.

Laboratory Tests

The SEM and EDAX analyses for clean freshly soldered tin/antimony solder are presented in Figures D-1 and D-2. Figure D-1 shows that the surface of the fresh solder is fairly smooth with only small surface irregularities and is free from a corrosion film. The EDAX analysis presented in Figure D-2 demonstrates that tin is the major constituent of the solder. Antimony does not appear on this diagram because it is masked by the much higher % composition of tin. Antimony is also difficult to distinguish from tin in the EDAX analysis because the Lα and Lβ lines for tin and antimony have similar values. Only the low intensity Lβ2 line of antimony is distinguishable from the tin lines. A listing of the principal emission lines from the elements in order of increasing energy can be found in Appendix D.

SEM photographs of the laboratory test coupons are presented in Figures D-3 and D-4. These coupons were held under standing water conditions for 48 hours, therefore only a thin passivation film is present.

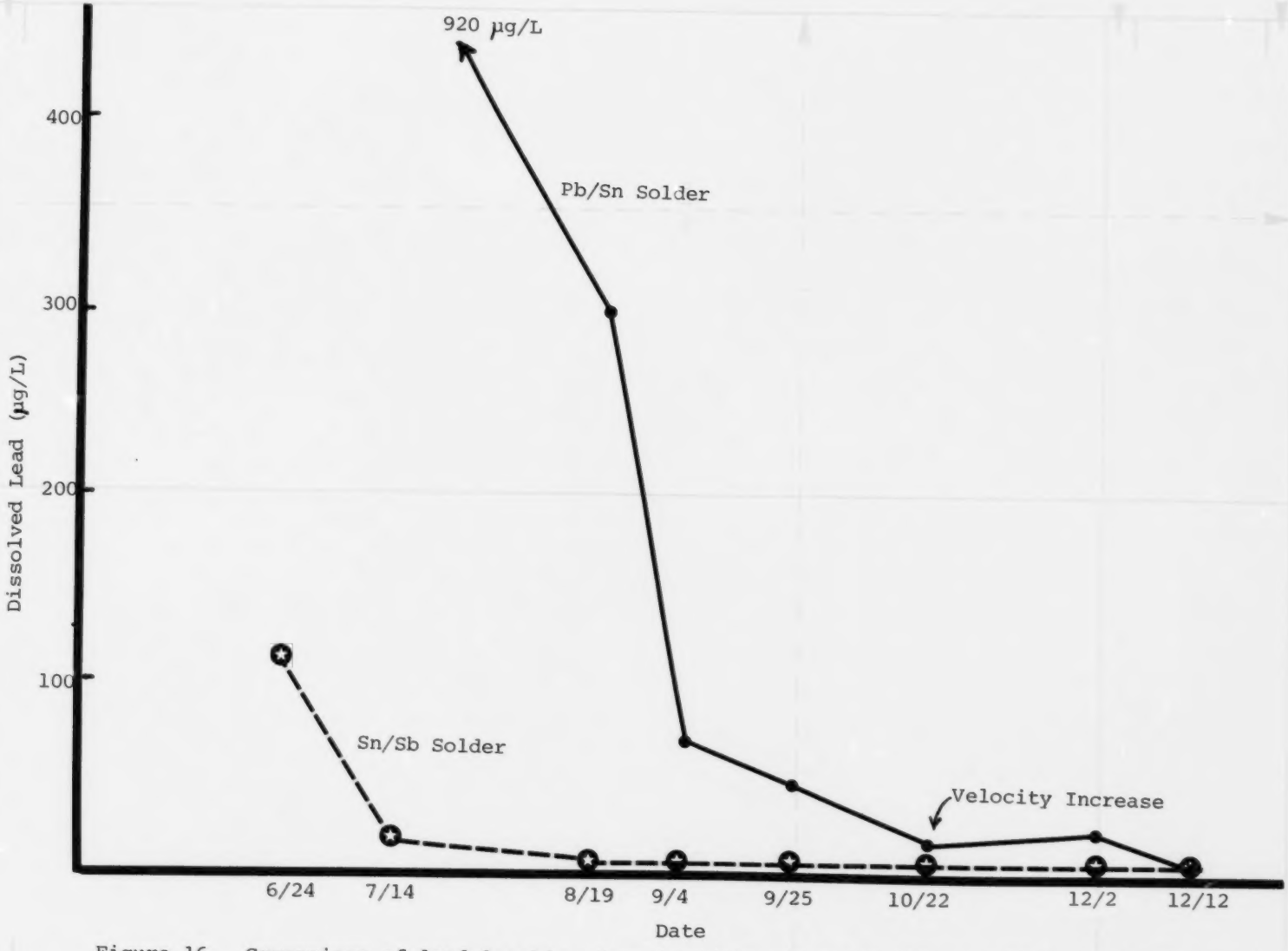

Figure 16. Comparison of lead leaching from lead/tin and tin/antimony solders (control loop).

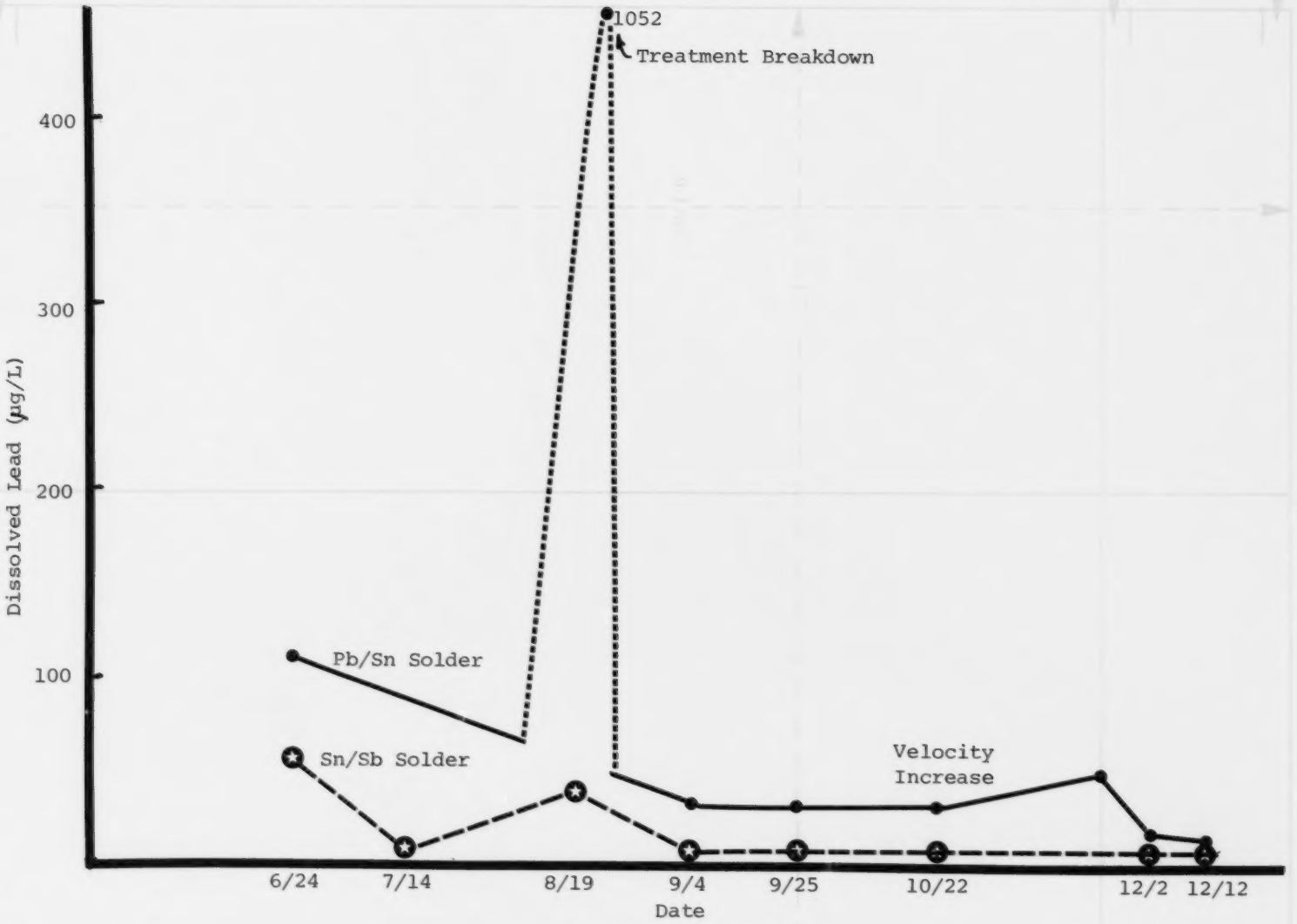

Figure 17. Comparison of lead leaching from lead/tin and tin/antimony solders (lime/sodium carbonate treatment).

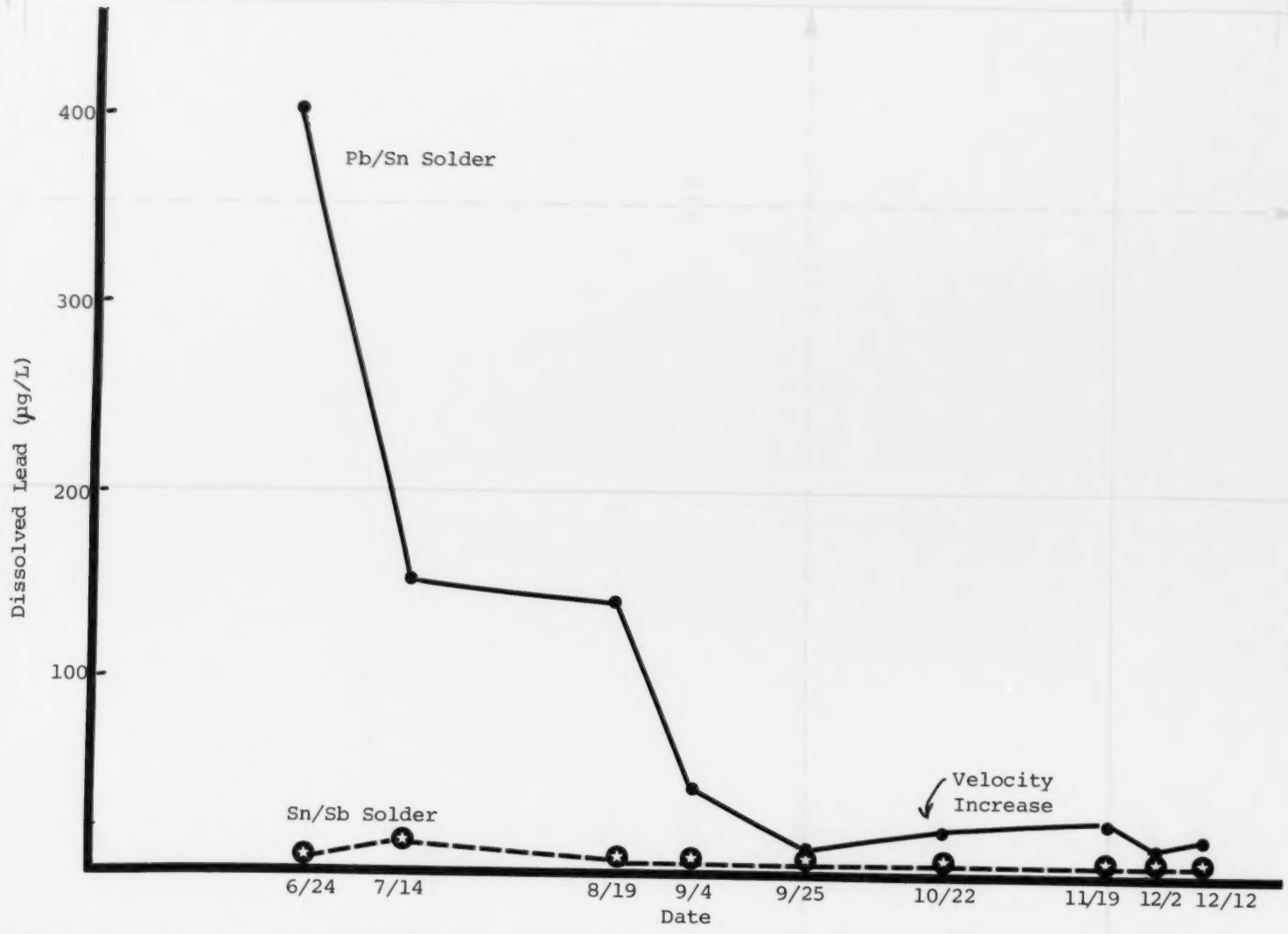

Figure 18. Comparison of lead leaching from lead/tin and tin/antimony solders (lime/bicarbonate treatment).

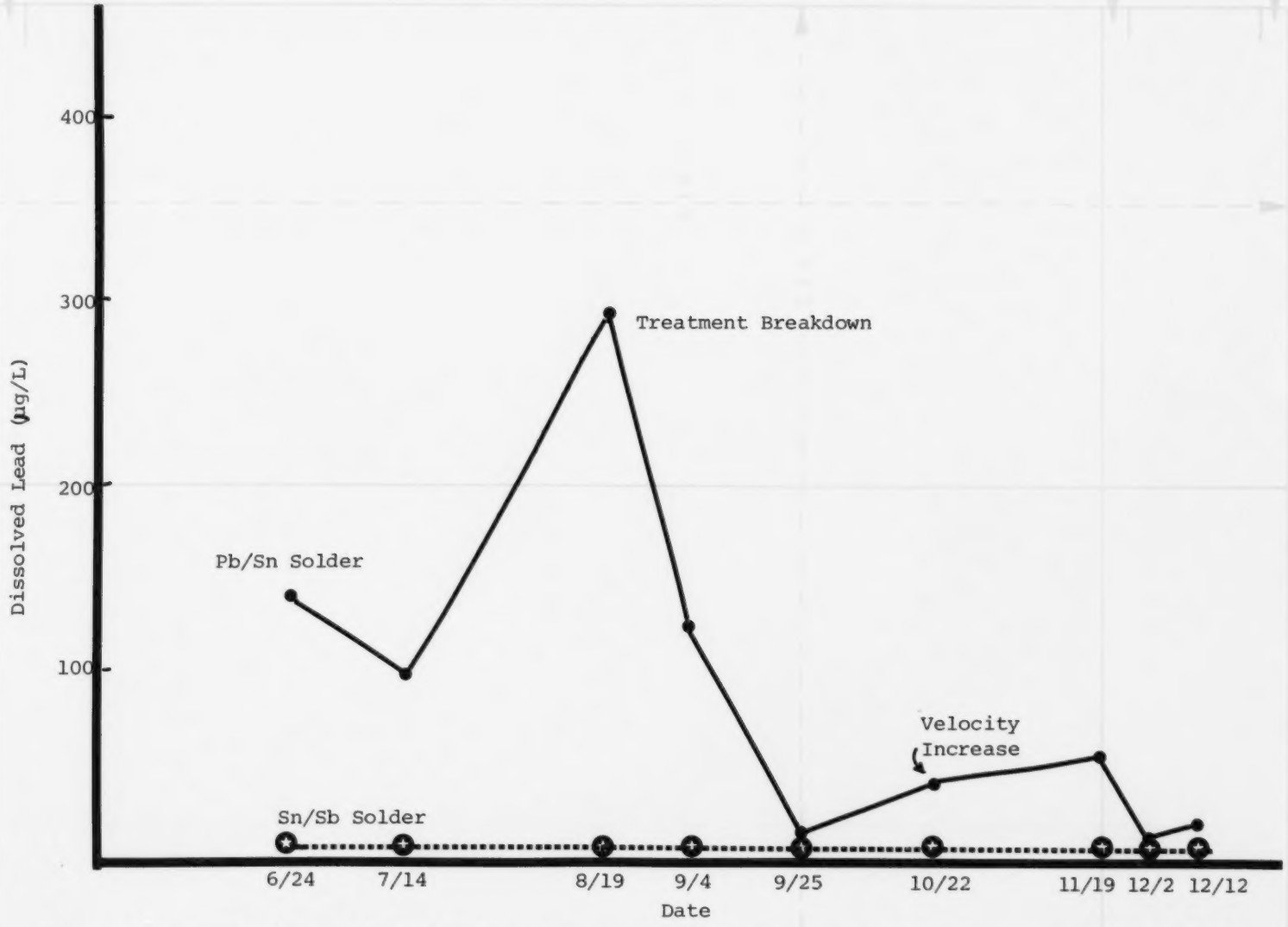

Figure 19. Comparison of lead leaching from lead/tin and tin/antimony solders (lime/bicarbonate/silicate treatment).

This film corresponds with a black colored film which was visible on the coupons.

Field Tests

Tin/antimony soldered joints were removed from buildings at the University of Washington and analyzed by SEM and EDAX. Figures D-5 and D-6 show at low magnification the two-layer protective film which forms on the solder. At higher magnification, the characteristics of the two layers are evident. The inner layer is dense, made up of very small, tightly packed grains (Figure D-7). The outer layer is composed of much larger particles which make it thick and porous (Figure D-8).

An elemental analysis of the two film layers was conducted and is presented in Figures D-9 and D-10. Figure D-9 demonstrates that the main constituent of the outer layer is tin. Copper, iron and silica are also present in large quantities. The composition of the inner film, presented in Figure D-10, again is mainly tin, with copper, iron, silica and antimony present in small amounts.

Metal Leaching Tests

Tin/Antimony Solder --

SEM and EDAX analyses were performed on the tin/antimony leaching test loop after 180 days exposure. Figures D-11 through D-14 demonstrate that the physical appearance of the film is not changed by increasing pH. In addition, the composition of the film is not significantly changed by treatment. The EDAX analysis presented in Figures D-15 through D-18 show that the main component of the film is tin,with trace quantities of silica, copper and iron. The silica content of lime/bicarbonate/silicate treatment loop corrosion film was somewhat greater than the control and the other treatment loops.

The similar characteristics of the corrosion film under control and treatment conditions, as analyzed with SEM and EDAX, agree well with the metal leaching test results. It can be concluded that because of the protective nature of the film under both control and treatment conditions, metal leaching will be minimal and is not affected by treatment.

The SEM analysis also pointed out some shortcomings in the metal leaching tests. Only three points of the solder are in direct contact with the copper pipe as illustrated in Figure 3. Near these three points there is a heavy corrosion film build-up (Figures D-11 through D-14) that becomes thinner away from the contact points (Figures D-19 and D-20). The uneven corrosion film results from severe galvanic corrosion near the copper contact points. This is not representative of actual plumbing systems, where the solder is in direct contact with copper on the entire piping joint.

SEM, EDAX and % composition analyses were also performed on the lead/tin solder leaching loop to determine why lead leaching in the control loop decreases with time to lead levels lower than the treatment loops. The SEM and EDAX analyses for the control, lime/soda ash, lime/bicarbonate, and

lime/bicarbonate/silicate treatment loops are presented in Figures D-21 through D-28. The film formed under the control conditions (Figure D-21) has a different physical appearance than the films formed under treatment conditions (Figures D-22, D-23 and D-24). The EDAX analysis shows that the major component of the film from the control loop is tin (Figure D-25), while in the treatment loops (Figures D-20, D-27 and D-28), lead is the major component. Therefore, in the control loop, it appears that the high level of lead leaching during the first few months of the test depletes the lead near the surface of the solder and a protective tin oxide film then forms. Contrastingly, lead is not depleted in the treatment loops but forms a lead oxide film on the surface of the solder. This lead film, can, however, dissolve and release lead into the drinking water if the pH conditions change suddenly. This is evidenced by the spikes which occurred following treatment breakdown in the metal leaching tests (see Figure 13).

Residential tap water monitoring in Seattle has also shown that lead continues to be leached from copper plumbing for many years after construction, thus indicating that the 50/50 lead/tin solder is not depleted of lead or passivated by a tin film under the changing flow conditions of a premise plumbing system. Therefore, it may be necessary to include on-off cycling in future metal leaching tests to better simulate actual flow conditions.

Summary of SEM and EDAX Analyses

The SEM and EDAX analyses demonstrated the physical and chemical characteristics of passivation films that formed on both tin/antimony and lead/tin solders. The film that forms on tin/antimony solder is composed of two layers - a thick, porous outer layer and an inner layer of small tightly packed grains. The major constituent of both layers in tin. On lead/tin solders, the chemical and physical characteristics of the corrosion film is dependent on pH. At low pH values, lead readily dissolves from the solder and as the solder ages, a tin passivation film can form. At high pH values, the solder surface can be passivated by a lead film.

SECTION 7

BENEFITS FROM USING TIN/ANTIMONY SOLDER

The theoretical, laboratory, field, and pilot results obtained in this study indicate that 95/5 tin/antimony solder has excellent potential for replacing lead-based solders in private and commercial plumbing systems. The use of tin/antimony solder in place of lead-based solders will result in lowered lead concentrations in tap water and therefore lower human lead exposure. Also, only minimal increases in antimony concentrations in drinking water are expected when tin/antimony solder is used, and when compared to the average antimony concentrations contained in the diet, drinking water concentrations are exceedingly small.

Tin/antimony solder is readily available and has been used with success in such places as the University of Washington. The tin/antimony solder, however, is about twice the cost of lead/tin solder. 1981 costs for a 2' length of solder are $3.00 and $1.55 for tin/antimony and lead/tin, respectively. The tin/antimony solder is also more difficult to work with because it has a smaller working temperature range.

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APPENDIX A

STANDARD FREE ENERGIES OF FORMATION

Formula	State	ΔG^o	Source
Sb	c	0	1
SbO^+	aq	-42.0	1
SbO_2^-	aq	-82.5	2
$HSbO_2$	aq	-97.5	2
Sb_4O_6	c	-298.0	1
Sb_2O_4	c	-165.9	2
Sb_2O_5	c	-200.5	1
SbO_3^-	aq	-122.9	3
SbO_2^+	aq	-65.5	3
SbH_3	g	35.3	2
SbF_3	c	-199.8	2
Sb_2S_3	amorph.	-32.0	2
$SbS_3^=$	aq	-32.0	2
SbS_2^-	aq	-13.0	2
$SbCl_3$	c	-77.62	1
F^-	aq	-66.08	1
HF	aq	-70.41	2
HF_2^-	aq	-137.5	2

(continued)

APPENDIX A (continued)

Formula	State	ΔG°	Source
Cl^-	aq	-31.35	1
Cl_2	aq	1.65	2
HCl	aq	-31.35	1
HClO	aq	-19.11	2
ClO^-	aq	08.9	2
$S^=$	aq	-21.96	3
HS^-	aq	3.01	1
H_2S	aq	-6.54	1
HSO_4^-	aq	-179.94	1
H_2SO_4	aq	-177.34	1
Sn	C	O	1
Sn^{2+}	aq	-6.275	2
Sn^{4+}	aq	0.65	2
SnO	C	-61.5	1
SnO_2	C	-123.2	3
$HSnO_2^-$	aq	-98.	2
SnS	C	-19.7	1
$Sn(SO_4)_2$	C	-346.8	2
$SnCl_2$	C	-72.2	2
$Sn(OH)^+$	aq	-60.6	3
$SnO_3^=$	aq	-137.42	3
SnH_4	g	99.	3

1. Rossini, F. D., D. D. Wagman, W. H. Evans, S. Levine, and I. Jaffe, Selected Values of Chemical Thermodynamic Properties: National Bureau Standards Circ. 500, U.S. Department of Commerce (1952).

(continued)

2. Latimer, W. M., Oxidation Potentials, 2nd Edition, New York, Prentice-Hall, 1952.

3. Technical Report 684, Enthapies Libre de Formation Standards, á 25°C. Centr Belee d'Étude de la Corrosion, Brussels, 1960.

APPENDIX B

THERMODYNAMIC EQUATIONS FOR AQUEOUS CORROSION OF ANTIMONY AND TIN

Table B-1

Thermodynamic Equations for Aqueous Corrosion of Antimony (Sb(s) - H_2O System) Figures 4 and 5

Equation	log K
1. $O_2(g) + 4H^+ + 4e^- = 2H_2O$	83.2
$pe = 20.8 - pH + 1/4 \log PO_2$	
2. $2H^+ + 2e^- = H_2(g)$	0
$pe = pH - 1/2 \log P_{H2}$	
3. $4Sb(s) + 6H_2O = Sb_4O_6 + 12H^+ + 12e^-$	-31.0
$pe = 2.6 - pH$	
4. $4SbO^+ + 2H_2O = Sb_4O_6(s) + 4H^+$	12.2
$pH = 3.1 - \log SbO^+$	
5. $4SbO_2^- + 4H^+ = Sb_4O_6(s) + 2H_2O$	56.9
$pH = 14.2 + \log SbO_2^-$	
6. $SbH_3(g) = Sb(s) + 3H^+ + 3e^-$	26.0
$pe = -pH - 8.7 + 1/3 \log P_{SbH_3}$	
7. $2Sb_2O_5(s) + 8H^+ + 8e^- = Sb_4O_6(s) + 4H_2O$	91.0
$pe = 11.4 - pH$	

(continued)

TABLE B-1 (continued)

Equation	log K
8. $2SbO_3^- + 2H^+ = Sb_2O_5(s) + H_2O$	8.4
$pH = 4.2 + \log SbO_3^-$	
9. $Sb_4O_6(s) + 6H_2O = 4SbO_3^- + 12H^+ + 8e$	-107.8
$pe = 13.5 - 3/2\ pH - 1/2 \log SbO_3^-$	
10. $4HSbO_2 = Sb_4O_6(s) + 2H_2O$	15.7
$\log HSbO_2 = -3.9$	
11. $SbO^+ + H_2O = HSbO_2 + H^+$	-0.9
$pH = 0.9 + \log [HSbO_2]/[SbO^+]$	
12. $HSbO_2 = H^+ + SbO_2^-$	-11.0
$pH = 11.0 + \log [SbO_2^-]/[HSbO_2]$	
13. $Sb(s) + 2H_2O = HSbO_2 + 3H^+ + 3e^-$	-11.7
$pe = 3.9 - pH + 1/3 \log HSbO_2$	
14. $Sb(s) + 2H_2O = SbO_2^- + 4H^+ + 3e^-$	-22.7
$pe = 7.6 - 4/3\ pH + 1/3 \log SbO_2^-$	
15. $Sb(s) + H_2O = SbO^+ + 2H^+ + 3e^-$	-10.8
$pe = 3.6 - 2/3\ pH + 1/3 \log SbO^+$	
16. $SbO^+ + 2H_2O = SbO_3^- + 4H^+ + 2e^-$	-23.9
$pe = 12.0 - 2pH + 1/2 \log [SbO_3^-]/[SbO^+]$	
17. $HSbO_2 + H_2O = SbO_3^- + 3H^+ + 2e^-$	-23.0
$pe = 11.5 - 3/2\ pH + 1/2 \log [SbO_3^-]/[HSbO_2]$	
18. $SbO_2^- + H_2O = SbO_3^- + 2H^+ + 2e^-$	-12.0
$pe = 6.0 - pH + 1/2 \log [SbO_3^-]/[SbO_2^-]$	

Table B-2

Thermodynamic Equations for Aqueous Corrosion of Antimony
($Sb - H_2O - S - Cl - F$ System)
(Figures 6 and 7)

Equation	log K
1. $O_2\ (g) + 4H^+ + 4e^- = 2H_2O$	83.2
$pe = 20.8 - pH + 1/2 \log PO_2$	
2. $2H^+ + 2e^- = H_2\ (g)$	0
$pe = -pH - 1/2 \log P_{H_2}$	
3. $SbO_3^- + 3HSO_4^- + 27H^+ + 25e^- = SbS_3^{-2} + 15H_2O$	161.5
$pe = 6.5 - 27/25\ pH - 1/25 \log SbS_3^{-2} + 1/25 \log - 3/25 \log HSO_4^-$	
4. $SbO_3^- + 3SO_4^{-2} + 30H^+ + 25e^- = SbS_3^{-2} + 15H_2O$	167.2
$pe = 6.7 - 6/5\ pH - 1/25 \log SbS_3^{-2} + 1/25 \log SbO_3^- - 3/25 \log HSO_4^-$	
5. $SbO^+ + 3HSO_4^- + 23H^+ + 23e^- = SbS_3^{-2} + 13H_2O$	137.6
$pe = 6.0 - pH - 1/23 \log SbS_3^{-2} + 1/23 \log SbO_3^- - 3/23 \log HSO_4^-$	
6. $HSbO_2 + 3HSO_4^- + 24H^+ + 23e^- = SbS_3^{-2} + 14H_2O$	138.5
$pe = 6.0 - 24/23\ pH - 1/23 \log SbS_3^{-2} + 1/23 \log SbO_3^- - 3/23 \log HSO_4^-$	
7. $HSbO_2 + 3SO_4^{-2} + 27H^+ + 23e^- = SbS_3^{-2} + 14H_2O$	144.2
$pe = 6.3 - 27/23\ pH - 1/23 \log SbS_3^{-2} + 1/23 \log SbO_3^- - 3/23 \log HSO_4^-$	
8. $SbO_2^- + 3SO_4^{-2} + 28H^+ + 23e^- = SbS_3^{-2} + 14H_2O$	155.3
$pe = 6.8 - 28/23\ pH - 1/23 \log SbS_3^{-2} + 1/23 \log SbO_3^- - 3/23 \log HSO_4^-$	
9. $2SbO^+ + 3HSO_4^- + 25H^+ + 24e^- = Sb_2S_3(s) + 14H_2O$	148.4
$pe = 6.2 - 25/24\ pH + 1/8 \log HSO_4^- + 1/12 \log SbO^+$	
10. $2HSbO_2 + 3HSO_4^- + 27H^+ + 24e^- = Sb_2S_3(s) + 16H_2O$	150.2
$pe = 6.3 - 9/8\ pH + 1/8 \log HSO_4^- + 1/12 \log HSbO_2$	

(continued)

TABLE B-2 (continued)

Equation	log K
11. $2HSbO_2 + 3SO_4^{=} + 30H^+ + 24e^- = Sb_2S_3(s) + 16H_2O$	155.2
$pe = 6.5 - 5/4\ pH + 1/8 \log HSO_4^- + 1/12 \log HSbO_2$	
12. $Sb_2S_3(s) + SO_4^{-2} + 8H^+ + 8e^- = 2SbS_2^- + 4H_2O$	31.9
$pe = 4.0 - pH - 1/4 \log SbS_2^- + 1/8 \log SO_4^{=}$	
13. $2SbS_2^- + 2H^+ = Sb_2S_3(s) + H_2S$	9.2
$pH = 4.6 - 1/2 \log H_2S + \log SbS_2^-$	
14. $Sb_2S_3(s) + 6H^+ + 6e^- = 2Sb(s) + 3H_2S$	-9.1
$pe = -1.5 - pH - 1/2 \log H_2S$	
15. $HSbO_2 + 2SO_4^{-2} + 19H^+ + 16e^- = SbS_2^- + 10H_2O$	-93.9
$pe = 5.9 - 19/16\ pH + 1/16 \log HSbO_2 + 2/16 \log SO_4^{-2}$	
16. $SbO_2^- + 2SO_4^{-2} + 20H^+ + 16e^- = SbS_2^- + 10H_2O$	104.9
$pe = 6.6 - 20/16\ pH + 1/16 \log SbS_2^- + 2/16 \log SO_4^{-2}$	
17. $SbS_2^- + 4H^+ + 3e^- = Sb(s) + 2H_2S$	0.06
$pe = .02 - 4/3\ pH - 2/3 \log H_2S + 1/3 \log SbS_2^-$	
18. $SbS_2^- + 2H^+ + 3e^- = Sb(s) + 2HS^-$	-14.0
$pe = -4.7 - 2/3\ pH - 2/3 \log HS^- + 1/3 \log SbS_2^-$	
19. $SbH_3(g) = Sb(s) + 3H^+ + 3e^-$	26.0
$pe = -pH - 8.7 + 1/3 \log P_{SbH_3}$	
20. $SbO^+ + H_2O = HSbO_2 + H^+$	-0.9
$pH = 0.9 + \log [HSbO_2]/[SbO^+]$	
21. $HSbO_2 = H^+ + SbO_2^-$	-11.0
$pH = 11.0 + \log [SbO_2^-]/[HSbO_2]$	

(continued)

TABLE B-2 (continued)

Equation	log K
22. Sb_4O_6 (s) + $2H_2O$ = $4HSbO_2$	-15.7
$\log HSbO_2 = -3.9$	
23. Sb_4O_6 (s) + $4H^+$ = $4SbO^+$ + $2H_2O$	-12.2
$pH = -3.1 - \log SbO^+$	
24. Sb_4O_6 (s) + $2H_2O$ = $4SbO_2^-$ + $4H^+$	-59.8
$pH = 15.0 + \log SbO_2^-$	
25. SbO^+ + $3Cl^-$ + $2H^+$ = $SbCl_3$ (s) + H_2O	-1.3
$pH = -0.6 + 3/2 \log Cl^- + 1/2 \log SbO^+$	
26. SbO^+ + $3F^-$ + $2H^+$ = SbF_3 (s) + H_2O	11.9
$pH = 6.0 + 3/2 \log F^- + 1/2 \log SbO^+$	

Table B-3

Thermodynamic Equations for Aqueous Corrosion of Tin
($Sn - H_2O - S - Cl$ System)
(Figures 8 and 9)

Equation	log K
1. $O_2(g) + 4H^+ + 4e^- = 2H_2O$ $pe = 20.8 - pH + 1/2 \log P_{O_2}$	83.2
2. $2H^+ + 2e^- = H_2(g)$ $pe = -pH - 1/2 \log P_{H_2}$	0
3. $Sn + H_2O = SnO + 2H^+ + 2e^-$ $pe = -pH - 1.76$	3.53
4. $Sn + 2H_2O = SnO_2 + 4H^+ + 4e^-$ $pe = -pH - 1.80$	7.19
5. $SnO + H_2O = SnO_2 + 2H^+ + 2e^-$ $pe = -pH - 1.84$	3.67
6. $Sn = Sn^{2+} + 2e^-$ $pe = 1/2 \log [Sn^{2+}] - 2.3$	4.60
7. $Sn^{2+} + 2H_2O = SnO_2 + 4H^+ + 2e^-$ $pe = -2pH - 1.3 - 1/2 \log [Sn^{2+}]$	2.60
8. $SnO_2 + H_2O = SnO_3^= + 2H^+$ $pH = 15.57 + 1/2 \log [SnO_3^=]$	-31.14
9. $Sn^{2+} = Sn^{4+} + 2e^-$ $pe = 2.54 + \log [Sn^{4+}] / [Sn^{2+}]$	-5.08
10. $Sn^{4+} + 2H_2O = SnO_2 + 4H^+$ $pH = -1.92 - 1/4 \log [Sn^{4+}]$	7.68

(continued)

TABLE B-3 (continued)

Equation	log K
11. $HSnO_2^- + H_2O = SnO_3^= + 3H^+ + 2e^-$	-12.66
$pe = -\frac{3}{2}pH - 6.33 + \log [SnO_3^=] / [HSnO_2^-]$	
12. $HSnO_2^- = SnO_2 + H^+ + 2e^-$	18.47
$pe = -1/2pH - 9.24 - 1/2 \log [HSnO_2^-]$	
13. $Sn + 2H2O = HSnO_2^- + 3H^+ + 2e^-$	-11.28
$pe = -\frac{3}{2} pH + 5.64 + 1/2 \log [HSnO_2^-]$	
14. $Sn^{++} + H_2O = Sn(OH)^+ + H^+$	-1.73
$pH = 1.73 + \log [Sn(OH)^+] / [Sn^{++}]$	
15. $Sn(OH)^+ + H_2O = SnO_2 + 3H^+ + 2e^-$	4.33
$pe = -3/2\ pH - 2.16 - 1/2 \log [Sn(OH)^+]$	
16. $Sn + H_2O = Sn(OH)^+ + H^+ + 2e^-$	2.87
$pe = -\ 1/2\ pH - 1.44 + 1/2 \log [Sn(OH)^+]$	
17. $SnCl_2 = Sn^{2+} + 2Cl^-$	50.57
$\log K = \log [Sn^{2+}] [Cl^-]^2$	
18. $SnS + 2H^+ = Sn^{2+} + H_2S$	-5.05
$pH = -2.53 - 1/2 \log [Sn^{2+}] [H_2S]$	
19. $SnS + H_2O + H^+ = Sn(OH)^+ + H_2S$	-6.78
$pH = -6.78 - \log [Sn(OH)^+] [H_2S]$	
20. $SnO_2 + H_2S + 2H^+ + 2e^- = SnS + 2H_2O$	-2.45
$pe = -pH + 1.23 + 1/2 \log [H_2S]$	
21. $Sn + H_2S = SnS + 2H^+ + 2e^-$	-9.65
$pe = -pH + 4.83 - 1/2 \log \frac{H_2S}{2}$	
22. $SnH_4(g) = Sn(s) + 4H^+ + re^-$	72.58
$pe = -pH - 18.16 - 1/4 \log P SnH_4$	

APPENDIX C

TABLE C-1. TEST LOOP QUALITY CONTROL (SUMMARIZED IN 2-WEEK INTERVALS)

	Loop #1, Control				Loop #2, Lime/Soda Ash			
Time Interval	Avg. pH	pH Range	Standard Deviation $\pm[H^+] \times 10^{-7}$	Tot. Alk. mg/L	Avg. pH	pH Range	Standard Deviation $\pm[H^+] \times 10^{-8}$	Tot. Alk. mg/L
1979 11/9-11/19	6.04	5.95-6.13	1.24	NA	7.38	6.58-8.80	5.32*	NA
11/19-11/29	6.13	6.08-6.29	1.05	NA	7.49	6.92-8.00	1.72	NA
1980 5/1-5/14	6.02	5.90-6.08	0.99	0.8	7.66	7.35-7.97	1.01	10.4
5/14-5/27	5.89	5.83-6.05	2.08	NA	7.60	7.19-8.50	1.29	NA
5/27-6/11	6.00	5.80-6.12	2.27	0.4	7.74	7.71-8.11	0.69	11.8
6/11-6/25	5.89	5.40-6.30	8.33	0.8	7.38	6.10-8.10	10.4*	6.4
6/25-7/9	6.07	5.92-6.18	0.90	0.9	7.60	7.35-8.69	1.29	10.6
7/9-7/23	5.80	5.20-6.15	13.8	2.2	7.50	6.60-8.05	3.62	1.27
7/23-8/6	5.94	5.80-6.18	2.46	NA	7.45	6.50-8.20	4.65*	NA
8/6-8/20	5.95	5.82-6.15	2.04	0.95	7.01	6.22-8.30	16.7*	9.1
8/20-9/3	6.07	5.99-6.22	1.38	NA	7.77	7.15-8.02	0.78	NA
9/3-9/17	6.05	5.92-6.12	1.24	1.4	7.69	7.30-8.11	0.75	6.4
9/17-10/1	6.12	6.02-6.20	0.85	1.5	7.61	6.38-8.55	5.33*	9.0
10/1-10/15	6.21	6.08-6.30	0.82	1.6	7.90	7.44-8.15	0.40	9.0
10/15-10/29	6.20	6.09-6.30	0.92	1.3	7.48	6.50-8.30	3.81*	7.6
10/20-11/12	6.09	5.94-6.21	1.82	-	7.73	7.40-8.02	0.36	NA

(continued)

TABLE C-1 (continued)

Time Interval	Loop #3, Lime/Bicarbonate Avg. pH	pH Range	Standard Deviation $\pm[H^+] \times 10^{-8}$	Tot. Alk. mg/L	Loop #4, Lime/Bicarbonate/Silicate Avg. pH	pH Range	$\pm[H^+] \times 10^{-8}$	Tot. Alk. mg/L
11/12-11/26	6.10	6.00-6.30	1.72	1.1	7.64	6.90-8.50	1.75	8.3
11/26-12/12	6.08	6.00-6.15	1.09	1.7	7.50	6.25-8.40	3.14*	11.5
1979 11/9-11/19	7.57	7.20-8.15	1.41	NA	7.67	7.40-7.97	0.90	NA
11/19-11/29	7.69	7.61-8.20	0.64	NA	7.75	7.32-8.02	0.64	NA
1980 5/1-5/14	7.58	7.20-8.07	2.98*	24.4	7.76	7.10-8.07	0.98	23.8
5/14-5/27	7.76	7.09-8.60	1.09	NA	7.69	7.21-8.26		NA
5/27-6/11	7.66	6.50-8.45	3.91*	18.6	7.91	7.30-8.70	0.60	20.6
6/11-6/25	8.01	7.65-9.40	0.33	13.4	7.89	7.71-8.25	0.87	15.6
6/25-7/9	7.92	7.40-8.35	0.45	16.2	7.95	7.31-8.72	0.68*	18.8
7/9-7/23		6.18-8.38		20.4	7.34	6.15-8.14	9.44*	20.6
7/23-8/6	7.74	6.99-8.40	1.53	NA	7.58	6.60-8.35	2.79*	NA
8/6-8/20	7.21	5.94-8.41	1.45*	18.5	7.74	6.75-8.32	1.55	17.0
8/20-9/3	7.53	6.70-8.40	3.01*	NA	7.87	7.01-8.24	1.16	NA
9/3-9/17	7.62	7.15-8.48	2.72*	9.4	7.82	7.10-8.15	0.90	9.6
9/17-10/1	7.42	6.40-8.24	5.09	16.8	7.55	6.75-8.21	2.45	16.4
10/1-10/15	7.93	7.60-8.48	0.72	15.7	7.90	7.10-8.42	1.05	16.2
10/15-10/29	7.83	7.20-8.51	0.76	19.2	7.89	7.10-8.65	0.54	15.3
10/29-11/12	7.84	7.45-8.13	0.37	NA	7.83	7.45-8.23	0.45	NA
11/12-11/26	7.69	7.30-8.50	1.36	18.8	7.85	7.25-8.50	0.86	NA
11/26-12/12	7.93	7.50-8.18	0.35	17.8	7.88	7.20-8.20	0.76	19.9

NA = Not analyzed.
*Upper standard deviation value not mathematically determinable.

TABLE C-2. METAL LEACHING TEST SECTIONS - FLOW AND VELOCITY SUMMARIZATION

Time Interval	Flow (L/sec)				Velocity, Copper Pipe (cm/sec)			
	Loop #1	Loop #2	Loop #3	Loop #4	Loop #1	Loop #2	Loop #3	Loop #4
1980								
5/22-6/24	6.59×10^{-3}	6.70×10^{-3}	6.42×10^{-3}	6.53×10^{-3}	13.4	13.6	13.0	13.3
6/26-7/14	6.63×10^{-3}	6.63×10^{-3}	6.51×10^{-3}	6.58×10^{-3}	13.5	13.5	13.2	13.4
7/16-8/10	6.57×10^{-3}	6.47×10^{-3}	6.58×10^{-3}	6.58×10^{-3}	13.3	13.1	13.4	13.4
8/21-9/4	6.53×10^{-3}	6.57×10^{-3}	7.64×10^{-3}	6.44×10^{-3}	13.3	13.3	13.5	13.1
9/6-9/25	6.67×10^{-3}	6.49×10^{-3}	7.41×10^{-3}	6.59×10^{-3}	13.5	13.2	13.0	13.4
9/27-10/22	6.57×10^{-3}	6.48×10^{-3}	6.65×10^{-3}	6.60×10^{-3}	13.3	13.2	13.5	13.4
10/24-11/19	1.00×10^{-2}	9.95×10^{-3}	9.97×10^{-3}	9.85×10^{-3}	20.3	20.2	20.2	20.0
11/23-12/2	1.02×10^{-2}	9.70×10^{-3}	1.01×10^{-2}	9.86×10^{-3}	20.7	19.7	20.5	20.0
12/4-12/12	9.84×10^{-3}	9.00×10^{-3}	1.01×10^{-2}	9.69×10^{-3}	20.0	18.3	20.5	19.7

(continued)

TABLE C-2 (continued)

Time Interval	Velocity Over 95/5 Tin/Antimony in Copper Pipe (cm/sec)				Velocity Over 50/50 Lead/Tin in Copper Pipe (cm/sec)			
	Loop #1	Loop #2	Loop #3	Loop #4	Loop #1	Loop #2	Loop #3	Loop #4
1980								
5/22-6/24	15.8	16.0	15.4	15.6	16.0	16.2	15.5	15.8
6/26-7/14	15.9	15.9	15.6	15.7	16.1	16.1	15.8	15.9
7/16-8/10	15.7	15.5	15.7	15.7	15.9	15.7	15.9	15.9
8/21-9/4	15.6	15.7	15.9	15.4	15.8	15.9	16.1	15.6
9/6-9/25	16.0	15.5	15.3	15.8	16.2	15.7	15.5	16.0
9/27-10/22	15.7	15.5	15.9	15.8	15.9	15.7	16.1	16.0
10/24-11/19	23.9	23.8	23.9	23.6	24.2	24.1	24.1	23.9
11/23-12/2	24.4	23.2	24.2	23.6	24.7	23.5	24.5	23.9
12/4-12/12	23.5	21.5	24.2	23.2	23.8	21.8	24.5	23.5

APPENDIX D

SCANNING ELECTRON MICROSCOPY AND X-RAY DIFFRACTION ANALYSIS

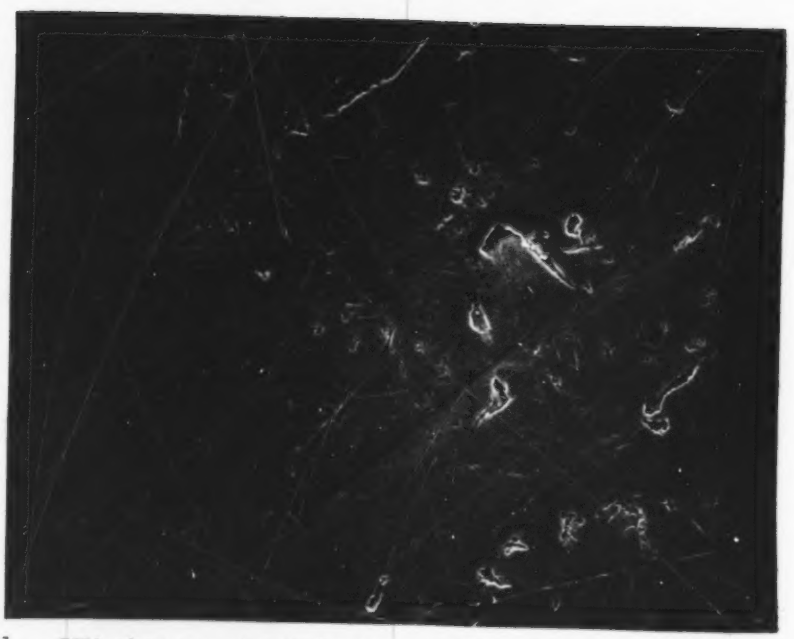

Figure D-1. SEM photograph of freshly soldered 95/5 tin/antimony solder. (Magnification = 534X)

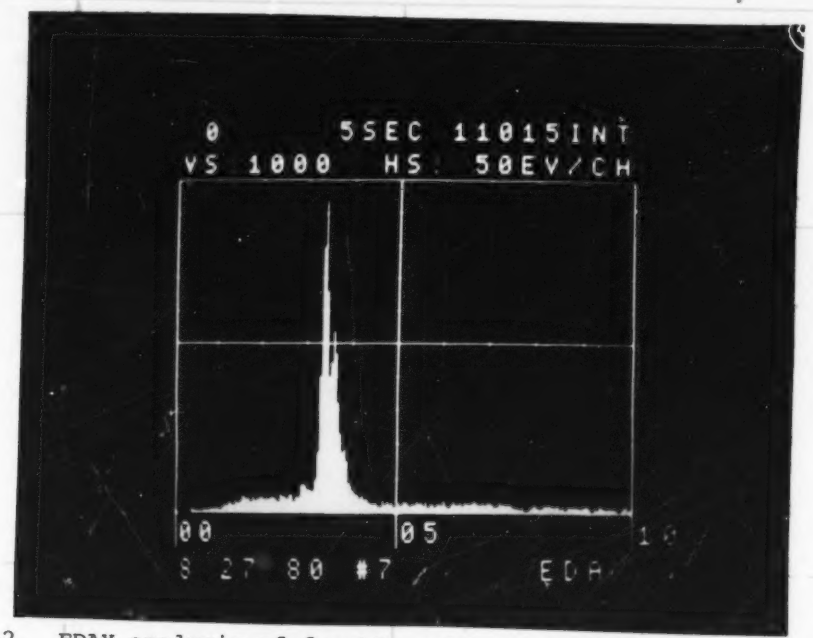

Figure D-2. EDAX analysis of freshly soldered 95/5 tin/antimony solder.

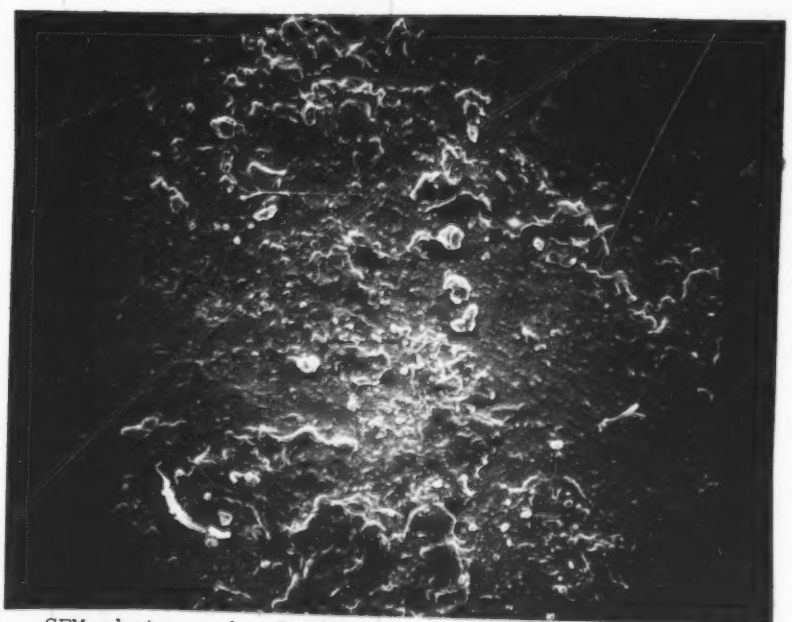

Figure D-3. SEM photograph of film growth on tin/antimony solder coupon. (Magnification = 240X)

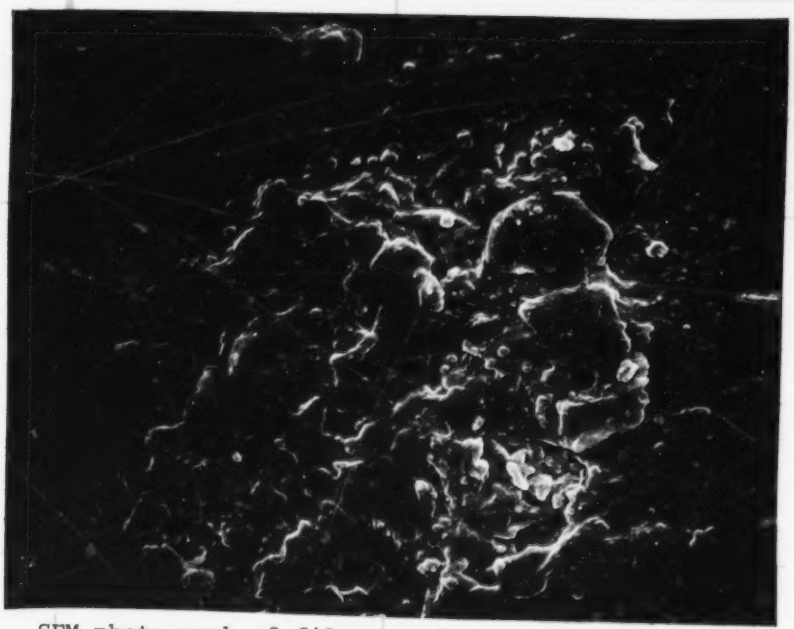

Figure D-4. SEM photograph of film growth on tin/antimony solder coupon. (Magnification = 600X)

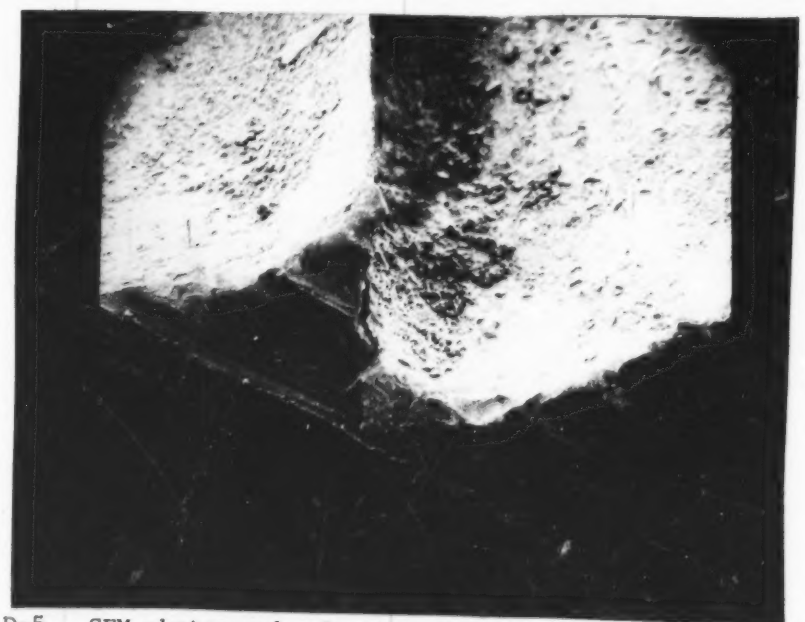

Figure D-5. SEM photograph of outer film growth on 95/5 tin/antimony soldered pipe joint. (Magnification = 23.5X)

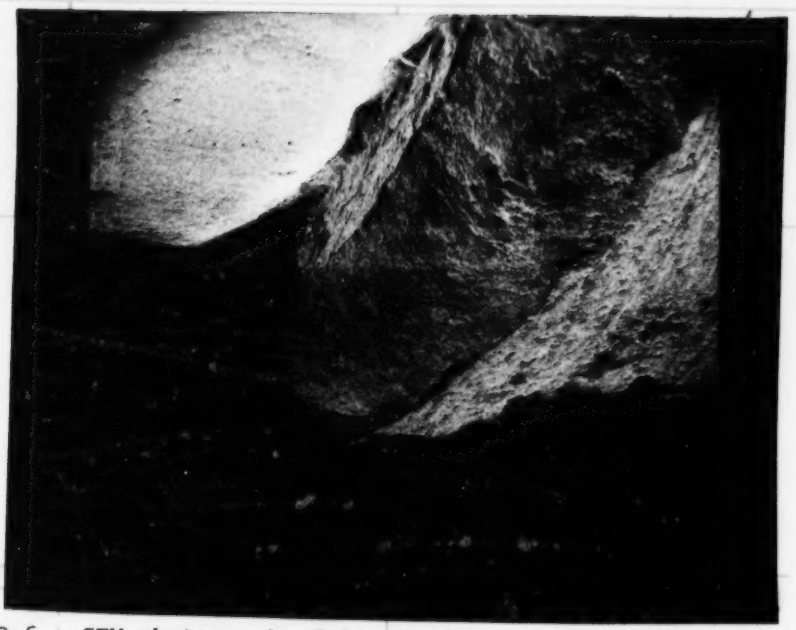

Figure D-6. SEM photograph of inner film growth on 95/5 tin/antimony soldered pipe joint.
(Magnification = 22.8X)

Figure D-7. SEM photograph of inner film growth on 95/5 tin/antimony soldered pipe joint.
(Magnification = 540X)

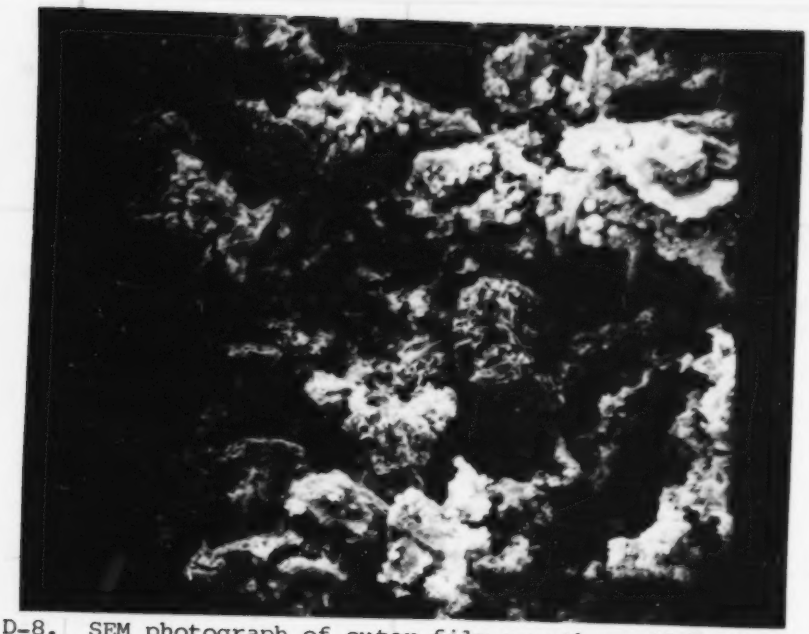

Figure D-8. SEM photograph of outer film growth on 95/5 tin/antimony soldered pipe joint.
(Magnification = 485X)

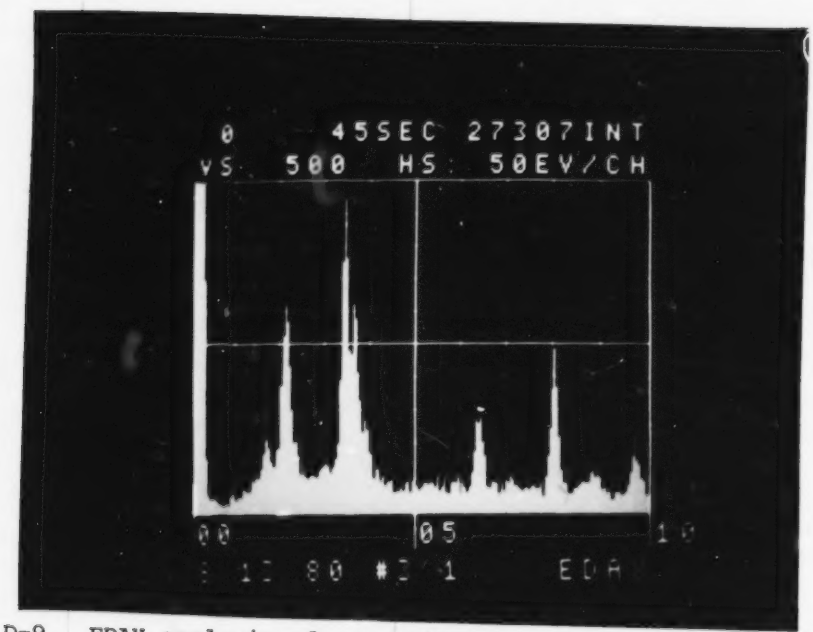

Figure D-9. EDAX analysis of outer film growth on 95/5 tin/antimony soldered joint.

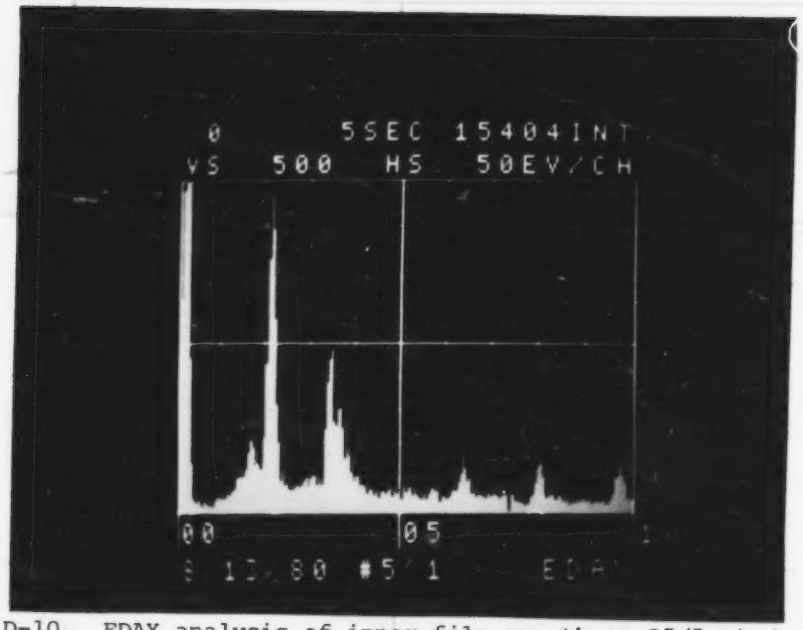

Figure D-10. EDAX analysis of inner film growth on 95/5 tin/antimony soldered joint.

Figure D-11. SEM photograph of corrosion film build-up on 95/5 tin/antimony solder control leaching section. (Magnification = 498X)

Figure D-12. SEM photograph of film build-up on 95/5 tin/antimony solder, lime/sodium carbonate treated leaching section. (Magnification = 486X)

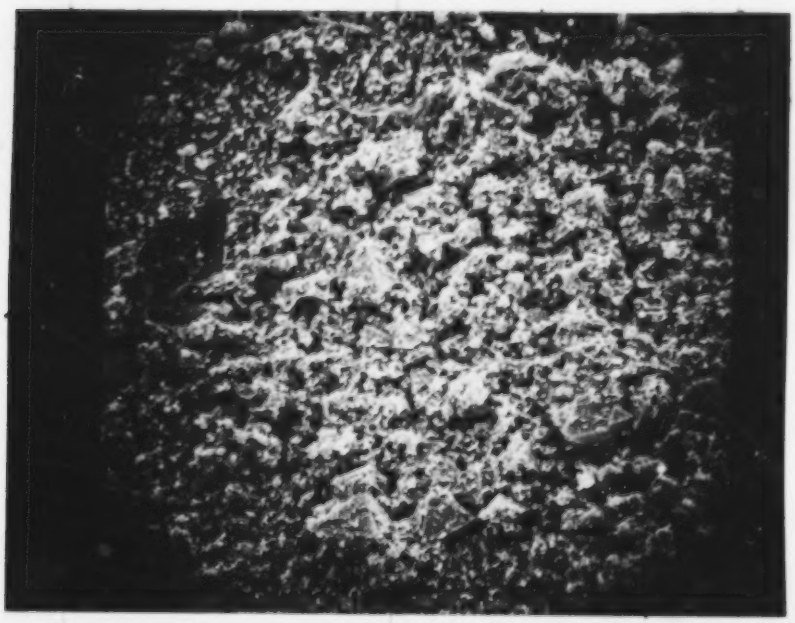

Figure D-13. SEM photograph of film build-up on 95/5 tin/antimony solder, lime/bicarbonate treated leaching section. (Magnification = 511X)

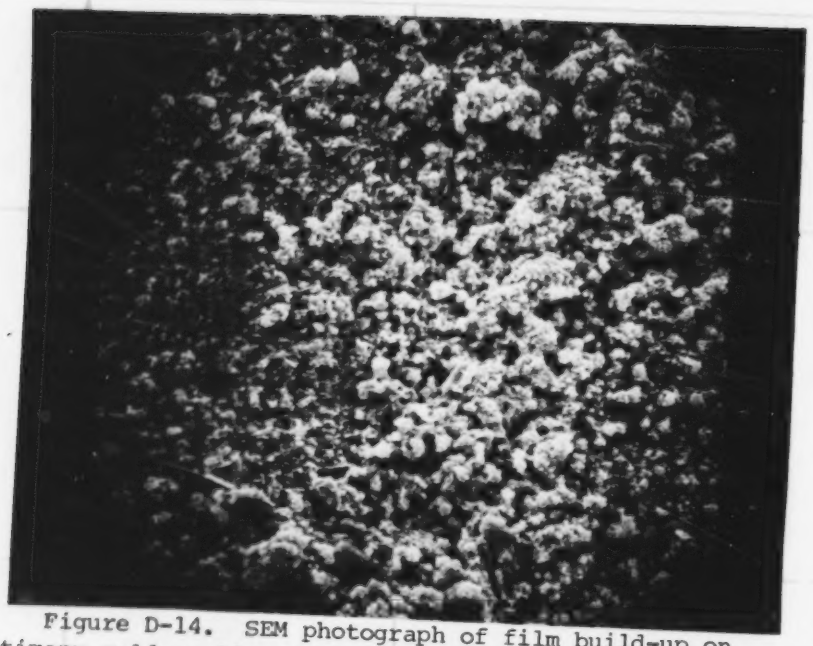

Figure D-14. SEM photograph of film build-up on 95/5 tin/antimony solder, lime/bicarbonate/silicate treated leaching section. (Magnification = 520X)

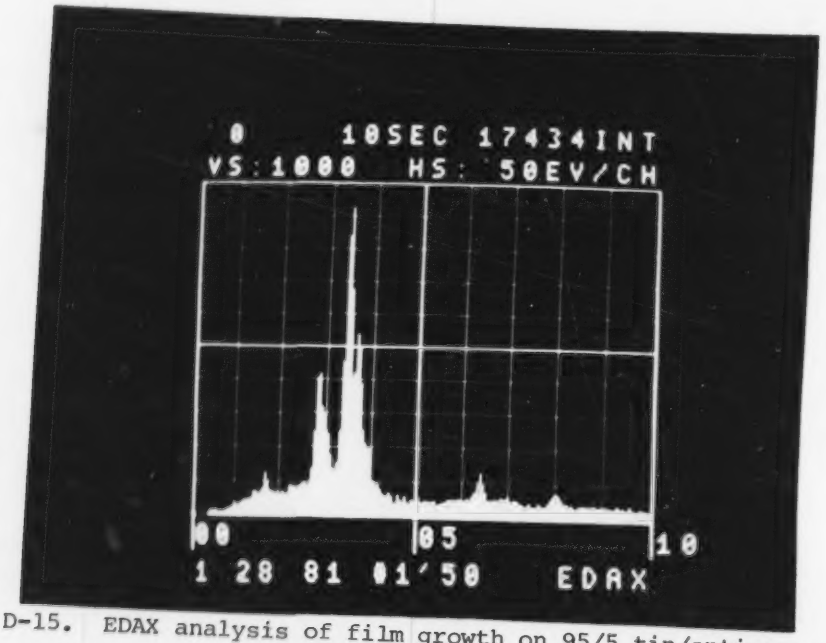

Figure D-15. EDAX analysis of film growth on 95/5 tin/antimony solder control leaching section.

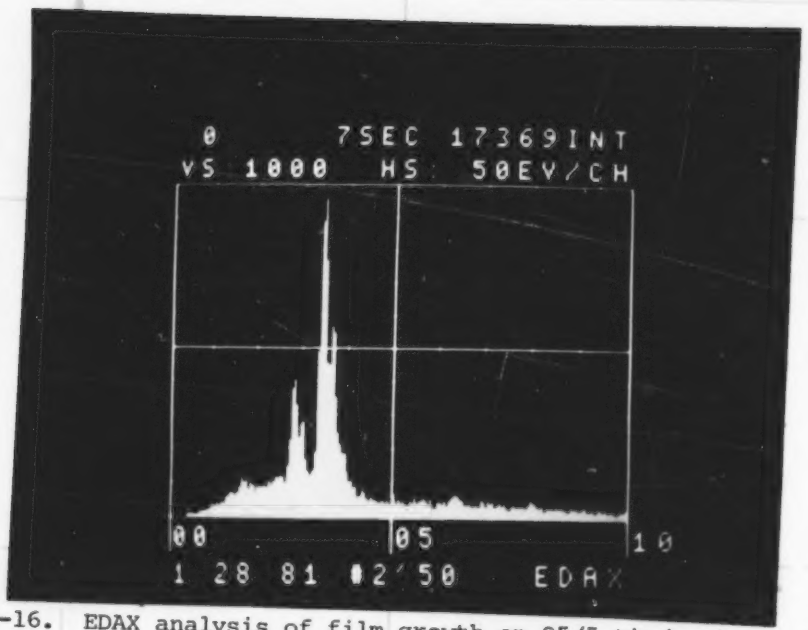

Figure D-16. EDAX analysis of film growth on 95/5 tin/antimony solder lime/sodium carbonate treated leaching section.

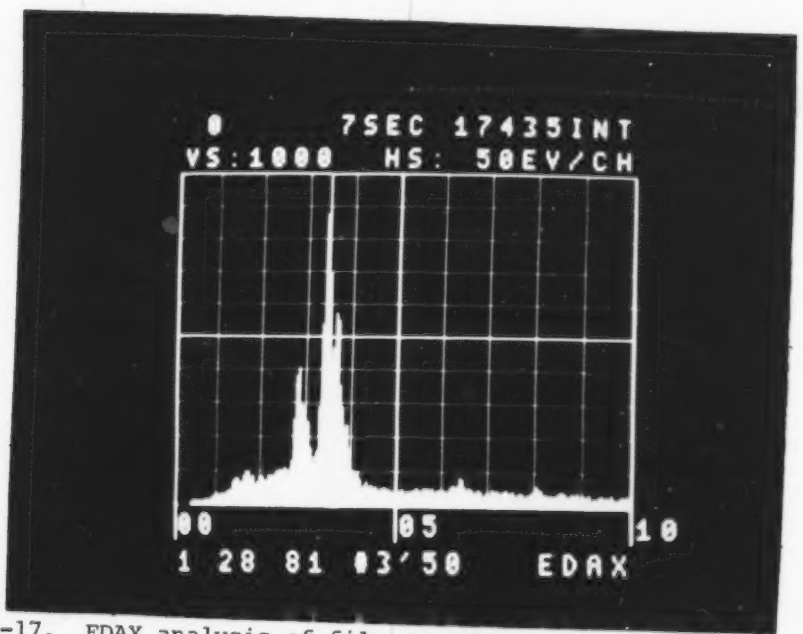

Figure D-17. EDAX analysis of film growth on 95/5 tin/antimony solder lime/bicarbonate treated leaching section.

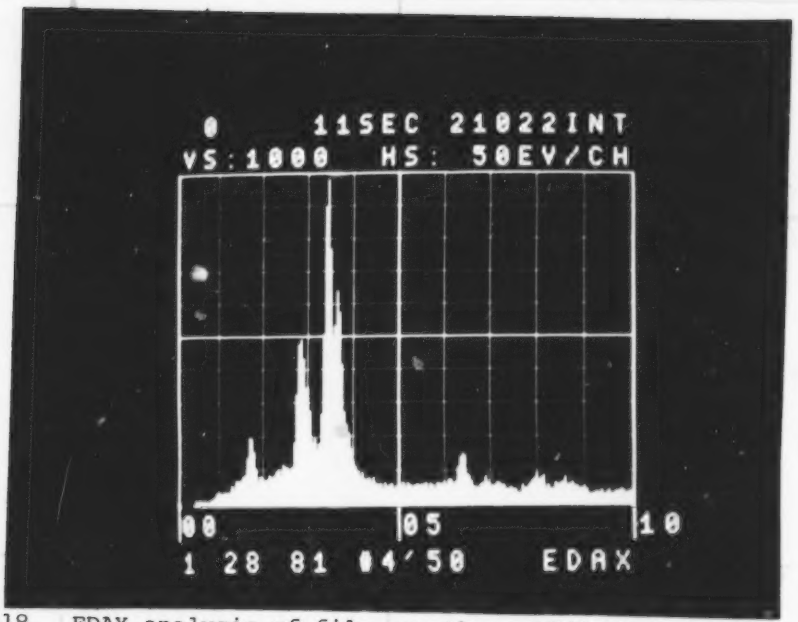

Figure D-18. EDAX analysis of film growth on 95/5 tin/antimony solder lime/bicarbonate/silicate treated leaching section.

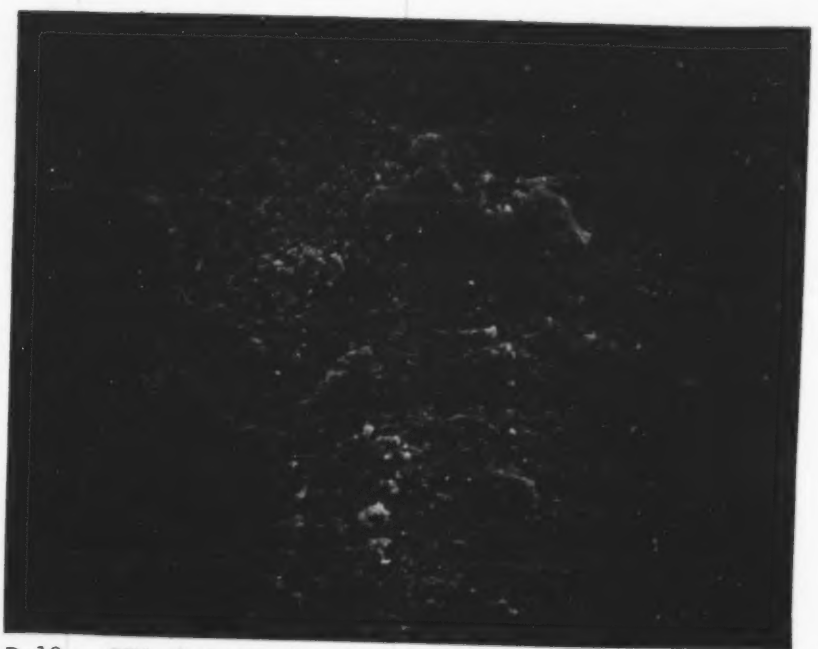

Figure D-19. SEM photograph of film formed on 50/50 lead/tin solder control leaching section.
(Magnification = 490X)

Figure D-20. SEM photograph of film formed on 50/50 lead/tin solder lime/sodium carbonate treated leaching section.
(Magnification = 524X)

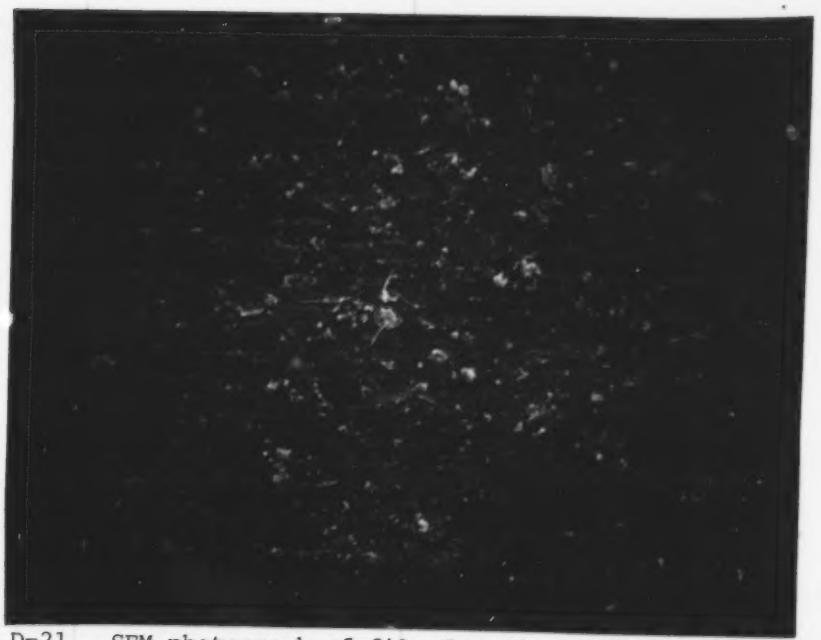

Figure D-21. SEM photograph of film formed on 50/50 lead/tin solder lime/bicarbonate treated leaching section.
(Magnification = 510X)

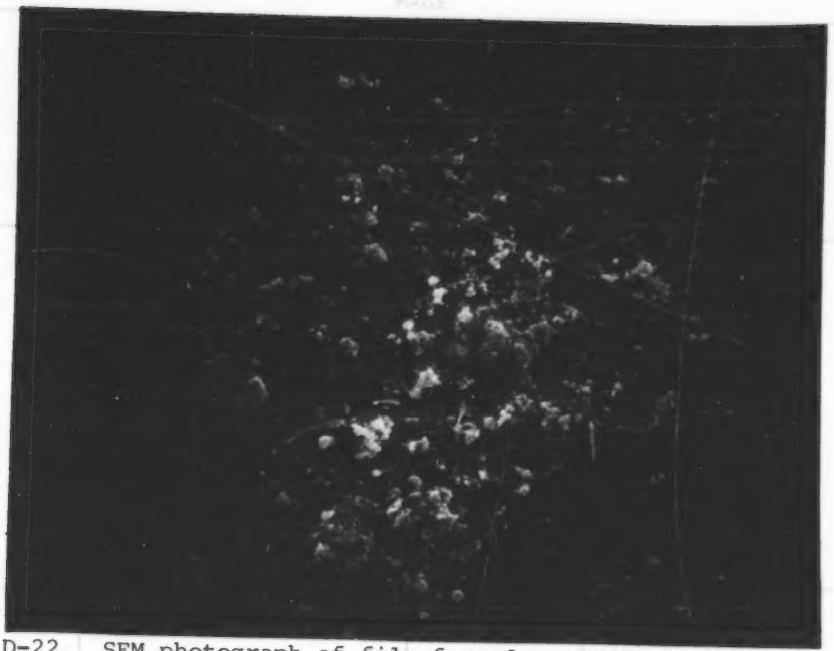

Figure D-22. SEM photograph of film formed on 50/50 lead/tin solder lime/bicarbonate/silicate treated leaching section. (Magnification = 510X)

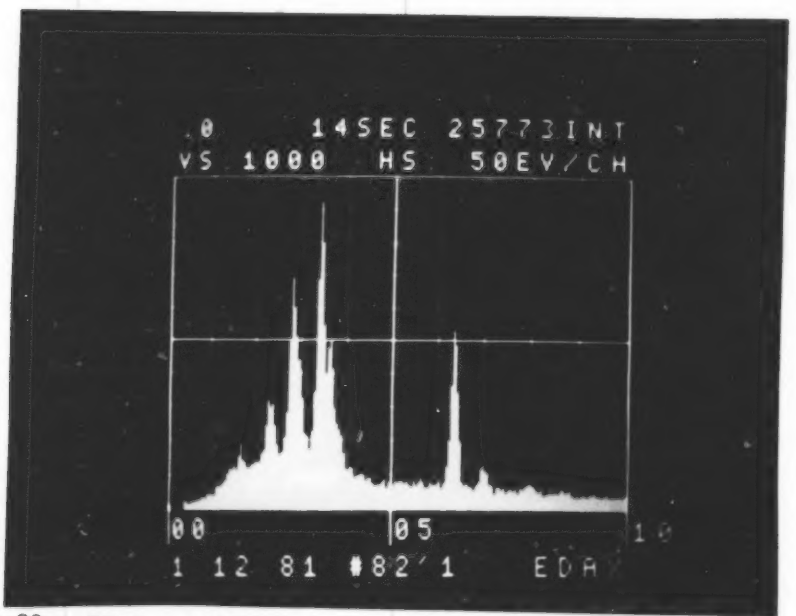

Figure D-23. EDAX analysis of film formed on 50/50 lead/tin solder control leaching section.

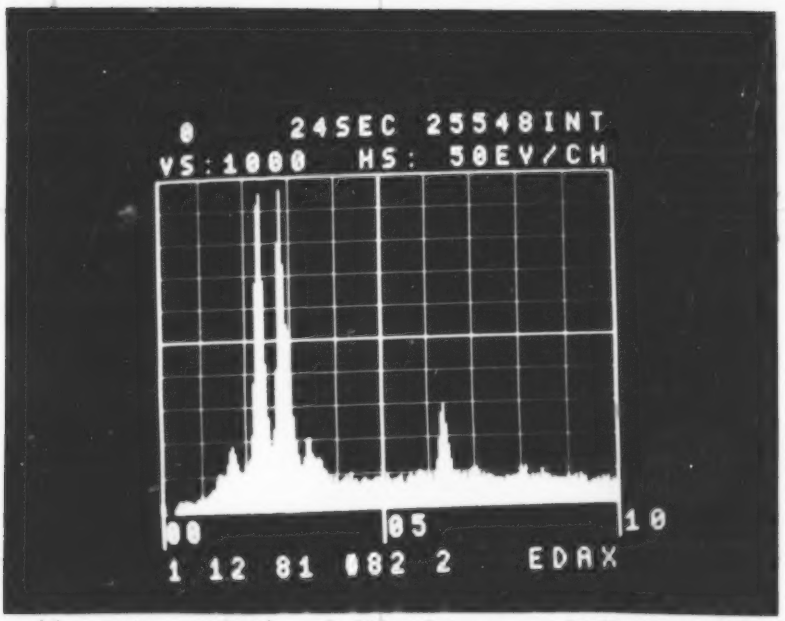

Figure D-24. EDAX analysis of film formed on 50/50 lead/tin solder lime/sodium carbonate leaching section.

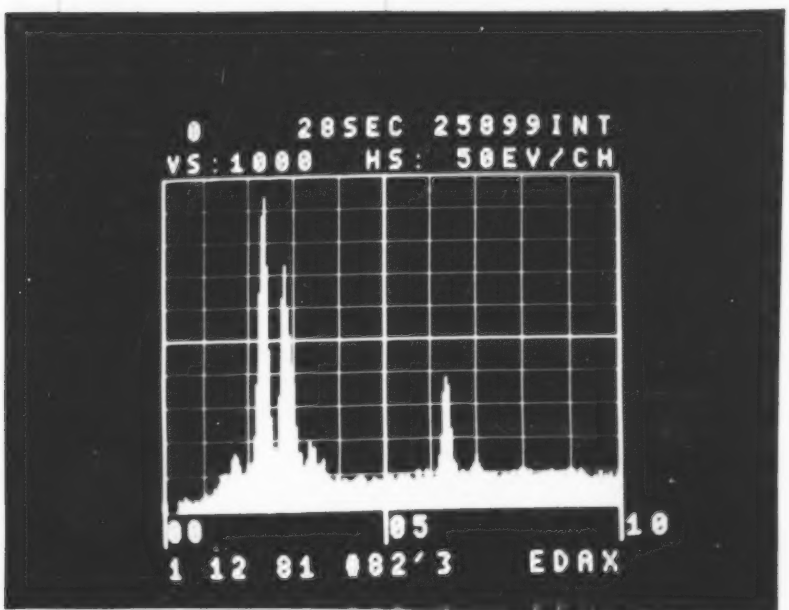

Figure D-25. EDAX analysis of film formed on 50/50 lead/tin solder lime/bicarbonate leaching section.

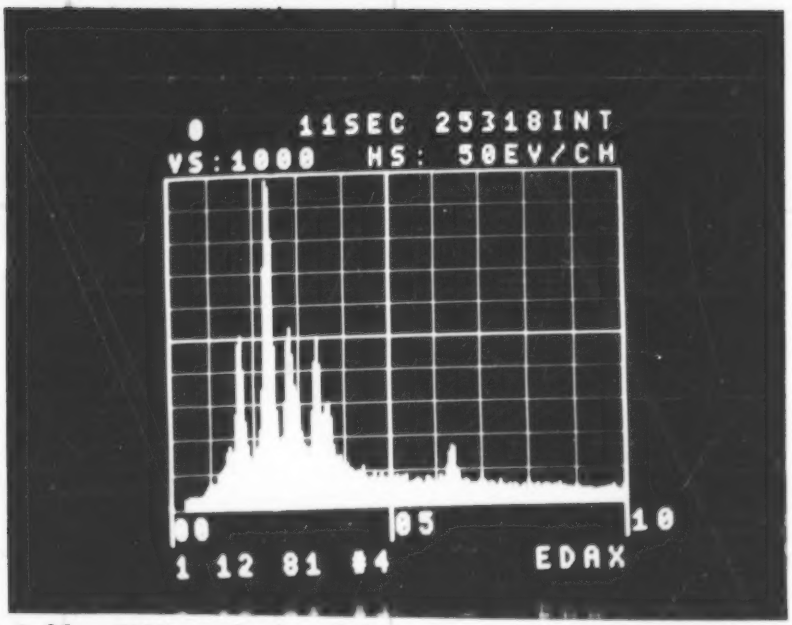

Figure D-26. EDAX analysis of film formed on 50/50 lead/tin solder lime/bicarbonate/silicate leaching section.

TABLE D-1. PRINCIPAL EMISSION LINES IN ORDER OF INCREASING ENERGY.

Energy	Element	Atmoic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
0-1 keV												
0.277	C	6	K									
0.331	Mo	42	M									
0.341	Ca	20	Lα									
0.355	Nb	41	Mγ									
0.392	N	7	K									
0.395	Sc	21	Lα									
0.452	Ti	22	Lα									
0.461	Ru	44	Mγ									
0.496	Rh	45	Mγ									
0.511	V	23	Lα									
0.525	O	8	K									
0.532	Pd	46	Mγ									
0.568	Ag	47	Mγ									
0.573	Cr	24	Lα									
0.606	Cd	48	Mγ									
0.637	Mn	25	Lα									
0.677	F	9	K									
0.691	Sn	50	Mγ									
0.705	Fe	26	Lα									
0.733	Sb	51	Mγ									
0.776	Co	27	Lα									
0.778	Te	52	Mγ									
0.833	La	57	Mα									
0.848	Ne	10	K									
0.851	Ni	28	Lα									
0.883	Ce	58	Mα									
0.929	Pr	59	Mα									
0.930	Cu	29	Lα									
0.972	Ba	56	Mγ									
0.978	Nd	60	Mα									

(continued)

TABLE D-1 (continued)

Energy	Element	Atomic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
1-2 keV												
1.012	Zn	30	Lα									
1.041	Na	11	Kα									
1.081	Sm	62	Mα									
1.098	Ga	31	Lα									
1.131	Eu	63	Mα									
1.185	Gd	64	Mα									
1.188	Ge	32	Lα									
1.240	Tb	65	Mα									
1.253	Mg	12	Kα									
1.282	As	33	Lα									
1.293	Dy	66	Mα									
1.347	Ho	67	Mα									
1.379	Se	34	Lα									
1.405	Er	68	Mα									
1.462	Tm	69	Mα									
1.480	Br	35	Lα									
1.486	Al	13	Kα									
1.521	Yb	70	Mα									
1.581	Lu	71	Mα									
1.586	Kr	36	Lα									
1.644	Hf	72	Mα									
1.694	Rb	37	Lα									
1.709	Ta	73	Mα									
1.739	Si	14	Kα									
1.774	W	74	Mα									
1.806	Sr	38	Lα									
1.842	Re	75	Mα									
1.914	Os	76	Mα									
1.922	Y	39	Lα									
1.977	Ir	77	Mα									

(continued)

TABLE D-1 (continued)

Energy	Element	Atomic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
2-3 keV												
2.013	P	15	$K\alpha_1$	2.028	$K\alpha_4$	(10)	2.137	$K\beta$	(6)			
2.042	Zr	40	$L\alpha_1$	2.124	$L\beta_1$	(45)						
2.048	Pt	78	$M\alpha$	2.127	$M\beta$	(50)						
2.121	Au	79	$M\alpha$	2.204	$M\beta$	(50)						
2.166	Nb	41	$L\alpha_1$	2.257	$L\beta_1$	(45)	2.163	$L\alpha_2$	(10)			
2.195	Hg	80	$M\alpha$	2.282	$M\beta$	(50)						
2.267	Tl	81	$M\alpha$	2.362	$M\beta$	(55)						
2.293	Mo	42	$L\alpha_1$	2.394	$L\beta_1$	(45)						
2.307	S	16	$K\alpha_1$	2.322	$K\alpha_4$	(50)	2.465	$K\beta$	(10)			
2.342	Pb	82	$M\alpha$	2.442	$M\beta$	(60)						
2.419	Bi	83	$M\alpha$	2.524	$M\beta$	(60)						
2.424	Tc	43	$L\alpha_1$	2.536	$L\beta_1$	(45)						
2.558	Ru	44	$L\alpha_1$	2.683	$L\beta_1$	(45)						
2.621	Cl	17	$K\alpha_1$	2.631	$K\alpha_2$	(10)	2.815	$K\beta$	(6)			
2.696	Rh	45	$L\alpha_1$	2.834	$L\beta_1$	(40)	3.001	$L\beta_2$	(25)			
2.838	Pd	46	$L\alpha_1$	2.990	$L\beta_1$	(40)	3.171	$L\beta_2$	(25)			
2.957	Ar	18	$K\alpha_1$	3.190	$K\beta$	(15)						
2.984	Ag	47	$L\alpha_1$	3.150	$L\beta_1$	(40)	3.347	$L\beta_2$	(25)			
2.991	Th	90	$M\alpha$	3.145	$M\beta$	(60)	3.369	$M\gamma$	(5)			
3-4 keV												
3.077	Pa	91	$M\alpha$	3.239	$M\beta$	(60)	3.465	$M\gamma$	(5)			
3.133	Cd	48	$L\alpha_1$	3.316	$L\beta_1$	(42)	3.528	$L\beta_2$	(25)			
3.165	U	92	$M\alpha$	3.336	$M\beta$	(60)	3.563	$M\gamma$	(5)			
3.286	In	49	$L\alpha_1$	3.487	$L\beta_1$	(75)	3.713	$L\beta_2$	(17)			
3.312	K	19	$K\alpha$	3.589	$K\beta$	(15)						
3.443	Sn	50	$L\alpha_1$	3.662	$L\beta_1$	(75)	3.904	$L\beta_2$	(17)			

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(continued)

TABLE D-1 (continued)

Energy	Element	Atomic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
3.604	Sb	51	$L\alpha_1$	3.843	$L\beta$	(75)	4.100	$L\beta_2$	(17)			
3.690	Ca	20	$K\alpha$	4.012	$K\beta$	(15)						
3.769	Te	52	$L\alpha$	4.029	$L\beta_1$	(75)	4.301	$L\beta_2$	(17)			
3.937	I	53	$L\alpha$	4.220	$L\beta_1$	(75)	4.507	$L\beta_2$	(17)			
4-5 keV												
4.088	Sc	21	$K\alpha$	4.460	$K\beta_1$	(20)						
4.109	Xe	54	$L\alpha$	4.42	$L\beta_1$	(50)	4.72	$L\beta_2$	(20)			
4.286	Cs	55	$L\alpha$	4.619	$L\beta_1$	(50)	4.935	$L\beta_2$	(20)			
4.465	Ba	56	$L\alpha$	4.827	$L\beta_1$	(50)	5.193	$L\beta_2$	(20)			
4.508	Ti	22	$K\alpha$	4.931	$K\beta_1$	(20)						
4.650	La	57	$L\alpha$	5.041	$L\beta_1$	(50)	5.383	$L\beta_2$	(20)			
4.839	Ce	58	$L\alpha$	5.261	$L\beta_1$	(50)	5.612	$L\beta_2$	(20)			
4.949	V	23	$K\alpha$	5.426	$K\beta_1$	(20)						
5-6 keV												
5.033	Pr	59	$L\alpha$	5.488	$L\beta_1$	(50)	5.849	$L\beta_2$	(20)			
5.229	Nd	60	$L\alpha$	5.721	$L\beta_1$	(50)	6.088	$L\beta_2$	(20)			
5.411	Cr	24	$K\alpha$	5.946	$K\beta_1$	(18)						
5.432	Pm	61	L	5.960	$L\beta_1$	(50)	6.338	$L\beta_2$	(20)			
5.635	Sm	62	$L\alpha$	6.204	$L\beta_1$	(50)	6.586	$L\beta_2$	(20)			
5.845	Eu	63	$L\alpha$	6.455	$L\beta_1$	(50)	6.842	$L\beta_2$	(20)			
5.894	Mn	25	$K\alpha$	6.489	$K\beta_1$	(20)						
6-7 keV												
6.056	Gd	64	$L\alpha$	6.712	$L\beta_1$	(50)	7.102	$L\beta_2$	(20)			
6.272	Tb	65	$L\alpha$	6.977	$L\beta_2$	(50)	7.365	$L\beta_2$	(20)			
6.398	Fe	26	$K\alpha$	7.057	$K\beta_1$	(20)						
6.494	Dy	66	$L\alpha$	7.246	$L\beta_1$	(50)	7.634	$L\beta_2$	(20)			

(continued)

TABLE D-1 (continued)

Energy	Element	Atomic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
6.719	Ho	67	Lα	7.524	$L\beta_1$	(50)	7.910	$L\beta_2$	(20)			
6.924	Co	27	Kα	7.648	$K\beta_1$	(20)						
6.947	Er	68	Lα	7.809	$L\beta_1$	(50)	8.188	$L\beta_2$	(20)			
7-8 keV												
7.179	Tm	69	Lα	8.100	$L\beta_1$	(50)	8.467	$L\beta_2$	(20)	9.424	$L\gamma_1$	(5)
7.414	Yb	70	Lα	8.400	$L\beta_1$	(50)	8.757	$L\beta_2$	(20)	9.778	$L\gamma_1$	(5)
7.471	Ni	28	Kα	8.263	$K\beta_1$	(20)						
7.654	Lu	71	Lα	8.708	$L\beta_1$	(50)	9.038	$L\beta_2$	(20)	10.142	$L\gamma_1$	(6)
7.898	Hf	72	Lα	9.021	$L\beta_1$	(50)	9.346	$L\beta_2$	(20)	10.514	$L\gamma_1$	(10)
8-9 keV												
8.040	Cu	29	Kα	8.904	$K\beta_1$	(20)						
8.145	Ta	73	Lα	9.342	$L\beta_1$	(50)	9.650	$L\beta_2$	(20)	10.893	$L\gamma_1$	(10)
8.396	W	74	Lα	9.671	$L\beta_1$	(50)	9.960	$L\beta_2$	(20)	11.284	$L\gamma_1$	(10)
8.630	Zn	30	Kα	9.570	$K\beta_1$	(20)						
8.651	Re	75	Lα	10.008	$L\beta_1$	(50)	10.274	$L\beta_2$	(20)	11.683	$L\gamma_1$	(10)
8.910	Os	76	Lα	10.354	$L\beta_1$	(50)	10.597	$L\beta_2$	(20)	12.093	$L\gamma_1$	(10)
9-10 keV												
9.174	Ir	77	Lα	10.706	$L\beta_1$	(50)	10.919	$L\beta_2$	(20)	12.510	$L\gamma_1$	(10)
9.241	Ga	31	Kα	10.262	$K\beta$	(21)						
9.441	Pt	78	Lα	11.069	$L\beta_1$	(50)	11.249	$L\beta_2$	(20)	12.940	$L\gamma_1$	(10)
9.712	Au	79	Lα	11.440	$L\beta_1$	(50)	11.583	$L\beta_2$	(20)	13.379	$L\gamma_1$	(10)
9.874	Ge	32	Kα	10.979	$K\beta$	(21)						
9.967	Hg	80	Lα	11.821	$L\beta_1$	(50)	11.922	$L\beta_2$	(20)	13.828	$L\gamma_1$	(10)

(continued)

TABLE D-1 (continued)

Energy	Element	Atomic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
10-11 keV												
10.267	Tl	81	Lα	12.211	$L\beta_1$	(50)	12.270	$L\beta_2$	(20)	14.289	$L\alpha_1$	(10)
10.542	As	33	Kα	11.722	Kβ	(22)						
10.550	Pb	82	Lα	12.612	$L\beta_1$	(50)	12.621	$L\beta_2$	(20)	14.762	$L\gamma_1$	(10)
10.837	Bi	83	Lα	13.021	$L\beta_1$	(50)	12.978	$L\beta_2$	(20)	15.245	$L\gamma_1$	(10)
11-12 keV												
11.129	Po	84	$L\alpha_1$	13.445	$L\beta_1$	(50)	13.338	$L\beta_2$	(20)	15.741	$L\gamma_1$	(10)
11.207	Se	34	Kα	12.492	Kβ	(24)						
11.425	At	85	$L\alpha_1$	13.574	$L\beta_1$	(50)	14.065	$L\beta_3$	(10)	16.249	$L\gamma_1$	(10)
11.725	Rn	86	$L\alpha_1$	14.313	$L\beta_1$	(50)	14.509	$L\beta_3$	(10)	16.768	$L\gamma_1$	(10)
11.907	Br	35	Kα	13.286	Kβ	(24)						
12-13 keV												
12.029	Fr	87	$L\alpha_1$	14.768	$L\beta_1$	(50)	14.448	$L\beta_2$	(20)	17.300	$L\gamma_1$	(10)
12.338	Ra	88	$L\alpha_1$	15.233	$L\beta_1$	(50)	14.839	$L\beta_2$	(20)	17.845	$L\gamma_1$	(10)
12.631	Kr	36	Kα	14.107	Kβ	(24)						
12.650	Ac	89	$L\alpha_1$	15.710	$L\beta_1$	(50)	15.929	$L\beta_3$	(10)	18.405	$L\gamma_1$	(10)
12.967	Th	90	$L\alpha_1$	16.199	$L\beta_1$	(50)	15.621	$L\beta_2$	(20)	18.979	$L\gamma_1$	(10)
13-14 keV												
13.288	Pa	91	$L\alpha_1$	16.699	$L\beta_1$	(50)	16.022	$L\beta_2$	(20)	19.565	$L\gamma_1$	(10)
13.373	Rb	37	Kα	14.956	Kβ	(24)						
13.612	U	92	$L\alpha_1$	17.217	$L\beta_1$	(50)	16.425	$L\beta_2$	(20)	20.164	$L\gamma_1$	(10)
13.942	Np	93	$L\alpha_1$	17.747	$L\beta_1$	(50)	16.837	$L\beta_2$	(20)	20.781	$L\gamma_1$	(10)

(continued)

TABLE D-1 (continued)

Energy	Element	Atomic Number	Principal Emission Line	Less Intense Minor Line			Lesser Intense Minor Line			Even Less Intense Minor Line		
				Energy	Line	Intensity	Energy	Line	Intensity	Energy	Line	Intensity
14-15 keV												
14.140	Sr	38	Kα	15.830	Kβ	(24)						
14.931	Y	39	Kα	16.734	Kβ	(25)						
15-16 keV												
15.744	Zr	40	Kα	17.663	Kβ	(27)						
16-17 keV												
16.581	Nb	41	Kα	18.700	Kβ	(12)						

APPENDIX E

SEATTLE WATER DEPARTMENT WATER ANALYSIS*

	Cedar			Tolt	
	†(1)	(2)	(3)	(4)	(5)
Alkalinity, Total (as $CaCO_3$)	20.5	21.0	20.0	6.4	2.1
Alkalinity, Bicarbonate (as $CaCO_3$)	20.5	21.0	20.0	6.4	2.1
Arsenic, Total, µg/l	<2	<2	<2	<2	<2
Barium	<.04	<.04	<.04	<.04	<.04
Cadmium, µg/l	.05	<.05	.05	.05	.05
Calcium (as $CaCO_3$)	18.5	22.5	21.2	7.6	8.8
Carbon Dioxide, free (calculated)	2.0	1.7	3.6	-	3.0
Chloride	0.75	1.9	3.3	0.8	2.6
Chromium	<0.15	<.015	<.015	<.015	<.015
Color, standard units	15	12	10	15	17
Copper	<.008	<.008	<.008	<.008	<.008
Fluoride	<0.10	0.98	1.00	<0.10	0.97
Hardness (as $CaCO_3$)(calculated)	22.4	27.2	26.0	9.3	10.3
Hardness, grains per gallon (calc.)	1.31	1.59	1.52	.54	.60
Iron	.045	.015	.015	.06	.065
Lead, µg/l	1	½	<½	<½	<½
Lithium, µg/l	0.2	0.3	0.25	<0.1	0.1
Magnesium	.94	1.15	1.16	.41	.37
Manganese, µg/l	3	2	2	5	<2
Mercury, Total, µg/l	-	-	<.1	-	<.1
Nitrate-Nitrogen	0.13	0.09	0.07	0.16	0.16
Ammonia-Nitrogen	<.01	<.01	<.01	<.01	<.01
Oxygen, Dissolved	10.28	9.58	9.99	10.15	9.54
Oxygen, % of saturation	98	92	98	97	96
pH	7.42	7.47	7.11	6.9	6.18
Phosphorus, Total Orthophosphate-PO_4	<.002	<.002	<.002	<.002	<.002
Potassium	.27	.27	.28	.19	.19
Residue, Total	47	50	51	26	30
Selenium, µg/l	-	-	<.005‡	-	<.005‡
Silica, Reactive	14.2	14.8	14.0	5.3	8.0
Silver, µg/l	<2	<2	<2	<2	<2
Sodium	1.73	1.77	1.77	.96	.96
Specific Conductance, µmhos	49.8	58.2	59.1	23.7	27.8
Strontium	.022	.026	.026	.009	.010
Sulfate	2.5	2.3	1.2	3.0	3.0
Tannin-Lignin (as Tannic Acid)	.20	<.03	<.03	.29	.16
Temperature, °C	13½	14	15	13½	16
Turbidity, NTU	0.7	0.35	0.4	0.5	0.45
Zinc, µg/l	<4	<4	<4	<4	<4

*Results given in mg/L except as noted.

†(1) Landsburg, Cedar River
(2) Lake Youngs near Intake
(3) Cedar Distribution at S. Forest Street & Airport Way S.
(4) Tolt Regulating Basin near Intake
(5) Tolt Distribution at N.W. 122nd Street & 1st Avenue N.W.

SAMPLES COLLECTED:
October 13, 1980

‡Analysis performed by Lauck's Testing Laboratories, Inc., Seattle, Washington.